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Research Triangle Park, North Carolina 27711

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16. ABSTRACT

The report gives results of a study of vanadia-alumina and iron oxide-chromium oxide-alumina catalysts for the reduction of NO with NH3 in simulated flue gas. Optimum catalyst compositions were 15% V2O5 on A12O3 and 10% Fe-Cr oxides on A12O3 with an Fe/Cr ratio of 1/9, respectively. Both catalysts were selective for the reduction of NO to N2 in the presence of O2. Both were shown to be sulfur-resistant during total periods of operation of 648 hours for V2O5 and 1052 hours for Fe-Cr in the presence of SOx. Typical conversion levels for V2O5 and Fe-Cr catalysts operating at 400 C in simulated flue gas containing 1000 ppm NO, 1000 ppm NH3, in the presence of SOx, were about 90 and 80%, respectively, at 20,000 per hour space velocity. Rate expressions for both catalysts have been developed for design use. A tabulation of recent publications in the field of NOx catalysis (particularly selective reduction with NH3) is presented.

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PARAMETRIC STUDIES OF CATALYSTS FOR NO_x CONTROL FROM STATIONARY POWER PLANTS

by

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SUMMARY

Chemical specifications of laboratory-prepared vanadium oxide-alumina and iron oxide-chromium oxide-alumina catalysts have been optimized for maximum activity for reduction of NO with NH_3 in simulated flue gas. Fifteen percent V_2O_5 loading and 10% Fe-Cr oxides with an Fe/Cr ratio of 9/1 were the optimum formulations.

A commercial 10% V_2O_5-Al_2O_3 catalyst was found to be almost as active as the 15% laboratory-prepared catalyst; no commercial Fe or Fe-Cr catalysts were found comparable to the prepared 9/1 Fe-Cr catalyst.

Parametric tests showed that both catalysts were selective in the presence of O_2 with strong enhancement in NO_x conversion rates due to the presence of O_2 under typical operating conditions. Neither CO_2 nor H_2O affect the NO_x reduction in the concentration ranges applicable to power plant exhaust. Both catalysts were most active between 400 and 425°C and required excess NH_3 ($NH_3/NO \geq 0.67$) for maximum activity.

Rate expressions describing the intrinsic reduction kinetics of NO with NH_3 in simulated flue gas over the V_2O_5 and Fe-Cr catalysts are:

V_2O_5-Al_2O_3:

$$\text{rate} = 2.05 \times 10^3 \, e^{-9650/RT} \, p_{NO}^{0.30} \, p_{NH_3}^{0.22} \, p_{O_2}^{0.05} \text{ (moles NO/gm. cat.-hr).}$$

10% Fe-Cr (9/1)-Al_2O_3:

$$\text{rate} = 3.25 \times 10^3 \, e^{-10,860} \, p_{NO}^{0.45} \, p_{NH_3}^{0.10} \, p_{O_2}^{0.15} \text{ (moles NO/gm. cat.-hr).}$$

Long-term durability tests in the presence of SO_x for periods of 648 and 1052 hours for V_2O_5 and Fe-Cr catalysts, respectively, have indicated no degradation in catalyst performance. In fact, improvement in activity and broadening of the conversion-temperature maxima were observed.

Typical conversion levels for V_2O_5 and Fe-Cr catalysts operating at 400°C in simulated flue gas containing 1000 ppm NO, 1000 ppm NH_3 and SO_x were about 90 and 80%, respectively, at 20,000 hr^{-1} space velocity.

With the Fe-Cr catalyst a sharp increase in the activity for reduction of low concentrations of NO_x (~ 500 ppm) was observed during the SO_x durability tests. With 500 ppm NO and stoichiometric NH_3 levels, conversions close to 90% at 20,000 hr^{-1} space velocity were typical with SO_x levels of about 350 ppm. With excess NH_3, NO_x conversions exceeding 90% were observed.

Two new catalysts were prepared in an attempt to reduce the cost of the V_2O_5 catalyst by dilution with other metal oxides. A 15% Fe-V (1/1) oxide-Al_2O_3 catalyst was prepared and preliminary tests indicate this catalyst to be more active than the 15% V_2O_5, the 10% Fe-Cr (9/1) and the 10% Fe_2O_3 catalysts. A second new catalyst, 10% Fe-Cr-V (18/1/1) oxide-Al_2O_3, was prepared during the last stages of this project and tests showed it to be the most active catalyst developed in the program. A test of about 180 hours duration of the latter catalyst in simulated flue gas containing SO_x at 400°C and 10,000 hr^{-1} showed that 99% removal of NO_x was achieved and activity did not deteriorate with exposure time. These preliminary results of the two catalysts suggest that synergistic catalytic activity has been achieved with mixtures of V_2O_5 and other metal oxides. Parametric and long term durability tests of these two new catalysts are recommended.

Although there was no deterioration of either the V_2O_5 or the Fe-Cr catalyst in simulated flue gas containing up to about 200 ppm SO_3, which was homogeneously formed from the injected SO_2, recent reports suggest that SO_3 may be the primary cause of the rapid deterioration of certain catalysts. The SO_3 levels or the ratio of SO_3 to SO_2 may have been too low to demonstrate this effect on the UCLA catalysts. Recent reports suggest interaction of SO_3 with (a) the active metal oxide and/or with (b) the Al_2O_3 carrier, respectively. Investigation of these possible interactions is of immediate importance. In particular, the influence of the SO_3/SO_2 ratio at various levels of total sulfur oxides should be studied. An SO_3 reaction with Al_2O_3 may require the development of SO_3-resistant carriers.

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1. INTRODUCTION

There has been considerable work on the physiological effects of the oxides of nitrogen, their role in the formation of smog and their production in fossil fuel-burning power generation sources. For many years research has been directed toward either preventing NO_x formation or after formation, its abatement.

Combustion modification methods have little effect on preventing the formation of NO_x from fuels containing nitrogenous compounds (1-1). The steadily increasing use of fuels containing such compounds (particularly coal) emphasizes the need for the development of post-combustion abatement methods.

A recent investigation performed by UCLA for TRW Systems Group under Environmental Protection Agency Contract No. 68-02-0648 (1-2), indicated that catalysis provides viable methods for controlling NO_x emissions. The most promising catalytic method is the selective reduction of NO_x in the presence of oxygen using ammonia as reductant. In brief, only ammonia has been identified as capable of reducing NO_x selectively in the presence of O_2; several catalysts have been developed that promote such selective reduction. During the TRW/UCLA project two catalysts were shown to be particularly promising. These catalysts were vanadium oxide (vanadia) - alumina and iron oxide - chromium oxide - alumina. In preliminary performance studies both catalysts were shown to be effective at flow rates and temperatures consistent with stationary power plant adaptability. Both were shown to be resistant to sulfur dioxide under short-term exposure.

In June 1975, UCLA initiated the present project (EPA Grant R 803653-01-0) to develop further these promising catalysts. The objectives of the project were to optimize the compositions of the vanadia and iron-chromium catalysts for selective reduction of NO_x with NH_3, develop rate expressions for design use and perform long-term durability studies of the optimum catalyst compositions in flue gas containing sulfur oxides.

Subsequent sections of this report summarize the previous effort and the experimental approaches and results of the current project. Immediately following is a description of the effort as delineated into separate study tasks.

CONCLUSIONS

The important conclusions based on the results of the current study are as follows:

1. Optimum loading of V_2O_5-Al_2O_3 catalysts, prepared by impregnation of Al_2O_3 with aqueous ammonium vanadate-oxalic acid for reduction of NO with NH_3, is 15% .
2. Optimum loading of Fe-Cr-Al_2O_3 catalysts, prepared by impregnation of aqueous nitrate mixtures on the carrier, is 10%. Optimum Fe/Cr ratio is 9/1.
3. Conversion of NO on both catalysts is not affected by the presence of H_2O, CO_2 or SO_x.
4. The presence of O_2 in the reactant gas mixture causes pronounced acceleration of the rate of NO reduction reaction on both catalysts.
5. Typical conversion of NO in simulated flue gas containing 1000 ppm NO and 1000 ppm NH_3 and SO_x was about 90% and 80% for V_2O_5 and Fe-Cr catalysts, respectively, at 400°C and 20,000 hr^{-1} space velocity. Lower space velocities enable attainment of higher NO conversion levels.
6. The rate of reduction of NO with NH_3 on 15% V_2O_5-Al_2O_3 catalyst in simulated flue gas up to 400°C can be described by the intrinsic rate expression,

   $$\text{rate} = 2.05 \times 10^3 \, e^{-9650/RT} \, p_{NO}^{0.30} \, p_{NH_3}^{0.22} \, p_{O_2}^{0.05} \quad \text{(moles NO/gm cat.-hr)}$$

7. The intrinsic rate expression for reduction of NO with NH_3 on 10% Fe-Cr (9/1)-Al_2O_3 in simulated flue gas is:

   $$\text{rate} = 3.25 \times 10^3 \, e^{-10,860} \, p_{NO}^{0.45} \, p_{NH_3}^{0.10} \, p_{O_2}^{0.15} \quad \text{(moles NO/gm cat.-hr)}$$

8. The V_2O_5 and Fe-Cr catalysts developed in this project suffered no degradation over extended periods of operation in simulated flue gas containing SO_x. In fact, some improvement in activity, particularly at low concentrations of

NO, was observed after extended SO_x exposure.

9. Both catalysts, which have been studied extensively in this project, require excess NH_3 for optimum operation. Residual NH_3 exists in the NO_x converter effluent. This residual NH_3 could present a secondary pollutant problem. No other secondary pollutants were found in the converter exhaust gases.

10. Although 15% V_2O_5 loading was optimum for V_2O_5-Al_2O_3 catalysts, more active catalysts can be prepared by substututing another active metal oxide for part of the vanadia. For instance, a 15% Fe-V (1/1) oxide on Al_2O_3 was more active than either 15% V_2O_5 or 10% Fe_2O_3. Also, a 10% Fe-Cr-V (18/1/1) oxide-Al_2O_3 was more active than either 15% V_2O_5 or 10% Fe-Cr (9/1).

11. Recent information suggests that the SO_3 in a SO_2-SO_3 mixture may be the primary cause of catalyst deterioration. Although the SO_x mixture was predominantly SO_2, moderate concentrations of SO_3 ($\leq$ 200 ppm) in simulated flue gas were always present during the current durability studies which showed no catalyst deterioration.

12. The nature of the possible catalyst degradation due to interaction with SO_3 is not clear. Other investigators suggest SO_3 reaction with active metals to form sulfates. However, interaction between SO_3 and Al_2O_3 carriers may be of greater significance. It is concluded that a more definitive effort in this area is of immediate importance.

RECOMMENDATIONS

The following recommendations are based on the results and conclusions of the current project:

1. The demonstrated activity and durability of 15% V_2O_5-Al_2O_3 and 10% Fe-Cr oxides (9/1) - Al_2O_3 at the bench scale of operation should be corroborated at larger (either semi-pilot or pilot) scale of operation using actual products of combustion of sulfur-containing fuels.

2. Recommended operating temperature for 15% V_2O_5 is 400°C and for 10% Fe-Cr (9/1) is 425°C. A space velocity of 10,000 hr^{-1} is recommended for initial performance evaluation at the larger scale.

3. Ammonia level should be maintained at a molar NH_3/NO ratio of 1 or higher for best NO_x removal.

4. Further investigation should be made of the variation in activity of V_2O_5 as a function of concentration of other active metal oxides and the imparted, apparent synergism. Parametric studies of these improved catalysts, particularly in the presence of varying SO_2/SO_3 mixtures is recommended.

5. Further studies of catalyst deterioration in the presence of larger concentrations of SO_3 than present in the current durability studies is of immediate importance. The SO_3-Al_2O_3 interaction should be examined carefully and the products or phase changes identified. Methods to improve the resistance of Al_2O_3 (additives, pretreatment, etc.) should be sought. Alternative carriers (e.g., zirconia) should be investigated to determine SO_3 resistance.

6. Studies should be made of methods to abate, recover and recycle, or utilize in other processes, the NH_3 remaining after NO_x abatement. The most attractive use of the NH_3 may be for SO_x abatement processes involving homogeneous reaction to form ammonium salts of sulfur oxides.

2. STATEMENT OF WORK

The experimental program performed in fulfillment of the objectives of Grant No. R 803653-01-0 "Parametric Studies of Catalysts for NO_x Control from Stationary Power Plants" was organized into the following tasks.

Task 1. Optimization of composition and development of chemical specifications for vanadia and iron-chromium catalysts.

Chemical composition and methods of preparation of vanadia and iron-chromium catalysts were correlated with catalytic activity for the selective reduction of NO with NH_3 in the presence of SO_x in simulated stack gas. Optimal concentrations of V_2O_5 on alumina and/or other carrier materials were determined. With the iron-chromium catalysts, the optimum ratio of Fe to Cr as well as the most active catalyst-carrier concentration ratio were determined. Commercial vanadia-based catalysts, currently used for oxidation of SO_2 to SO_3, in nitric acid production and in catalytic stack gas desulfurization processes, were investigated for activity for NO reduction with NH_3.

Task 2. Parametric investigation of vanadia and iron-chromium catalysts in the presence of SO_2.

Process variables such as temperature and residence time were varied over broad ranges with the optimized vanadia and iron-chromium catalyst formulations. The effect of SO_2 concentration on the temperature-activity characteristics was determined. Tests of long duration were made to determine if slow, cumulative poisoning occurs. Working rate expressions were developed for scale-up use. Analyses of the effluent gas stream determined not only the concentrations of the major components of interest (e.g., NO, NO_2, NH_3, SO_2, SO_3) but also those of other components which may represent deleterious secondary effects (e.g., sulfates, H_2S, carbonyls).

Task 3. Investigation of new and promising catalysts.

Promising catalysts were tested when the literature or informal disclosures showed evidence of high activity. Following preliminary assessment of these catalysts based on available data, recommendations for further investigation were made to the EPA and pursued, if merited, upon the directive of the project officer.

3. BACKGROUND AND DESCRIPTION OF THE PROBLEM

Several approaches considered for abatement of NO_x in the exhaust of fossil fuel-burning power plants involve catalysis. These approaches may be summarized as:

1. Catalytic Decomposition of NO

Nitric oxide is formed in combustion processes by direct combination of nitrogen and oxygen at high temperature. The equilibrium concentration of NO is determined by the flame temperature which depends on the air to fuel ratio. Thermodynamically, nitric oxide should revert to nitrogen and oxygen as the combustion gases cool. However, the kinetics of the homogeneous NO decomposition reaction become infinitesimally slow as the gases are cooled and the NO concentration in the flue gas is essentially that in the combustion zone. Catalysts should exist that will accelerate the approach to equilibrium (lower compositions of NO) as the gases are cooled, at least until temperatures become so low that kinetic restraints exist for the heterogeneous process. Some success was achieved in identification of such a catalyst for the direct decomposition of NO in the previous program.* The catalyst was platinum-alumina which, under typical flue gas conditions, resulted in 60% decomposition of NO. The moderate conversion attained and the high cost of platinum indicate that continued development of this process is only marginally attractive at the present time.

2. Nonselective Reduction of NO

It has been known for many years (e.g. tail-gas treatment operations in the nitric acid industry) that the reduction of NO can be catalytically accomplished by addition of sufficient reducing agent to reduce both NO and oxygen. Typical reducing agents (H_2, CH_4, CO, gasoline, etc.) will effectively reduce NO in the absence of oxygen or, in the presence of oxygen if there is an excess of reducing agent to reduce both the O_2 and the NO. For conditions of substantial excess O_2, (typically, under optimum fuel-utilization conditions in a power plant), the heat release in the preliminary O_2-depletion reaction is such that intercooling and multistage reactors are necessary.

Nonselective reduction of NO presents two alternative requirements. Either the combustion process must be conducted under uneconomical O_2-deficient conditions (which also leads to lower formation of NO) or the reducing agent must be provided from external sources. For this latter alternative, sufficient reductant must be added to consume also the O_2 as described above.

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* Reference will be made in this report to the joint TRW/UCLA effort recently concluded (1 -2) as the previous program.

There are many catalysts which will simultaneously reduce both NO and O_2, but none has been identified that will promote the selective reduction of NO using standard fuels, over practical operating ranges, in the presence of excess O_2. In summary, non-selective reduction of NO is technically feasible for power plant NO abatement but the economic shortcomings of the alternative of fuel-rich combustion conditions or complicated reactor/intercooling systems with large excess reductant requirements make such systems relatively unattractive.

3. Catalytic Oxidation of NO

Although thermodynamic analysis indicate that NO should readily decompose to N_2 and O_2 at lower temperatures, the reaction is kinetically slow. The formation of NO_2 from NO and O_2 is also favored as the combustion gases cool. However, the low residence times inherent in power plant exhaust systems limit the degree of oxidation.

If the oxidation process can be accelerated by catalysis, removal of the more active NO_2 in secondary scrubbing processes is feasible. However, no heterogeneous catalyst has been identified as yet that will promote the oxidation process to a significant extent under typical stack-gas conditions.

4. Selective Reduction of NO

Only one reducing agent, NH_3, has been found that will react selectively with NO in the presence of large amounts of O_2 over wide range of reductant/NO mixture ratios. During the previous project it was found that platinum, vanadia and iron oxide-chromium oxide mixtures on alumina support material were effective catalysts for the selective reduction of NO with NH_3. The latter two catalysts were disclosed in the patent literature by Nonnenmacher and Kartte (3-1) and by Schmidt and Schulze (3-2), respectively.

Samples of each of the three catalysts, $Pt-Al_2O_3$ (from Engelhard), $V_2O_5-Al_2O_3$ (from Harshaw) or Fe-Cr oxide-Al_2O_3 (laboratory prepared) were subjected to a brief parametric investigation to study the effects of NH_3/NO ratio, NO, O_2 and SO_2 concentrations, space velocity (s.v.) and temperature on the conversion of NO. It was found that the non-noble metal catalysts promoted the selective reduction of NO with a maximum in conversion of NO at 400°C for 20,000 hr^{-1} space velocity (STP). Although the platinum catalyst showed a conversion maximum at only 250°C, a major product of the reaction was N_2O. On the other hand, the non-noble metal catalysts did not promote extensive production of N_2O. As space velocity increased, conversion of NO on both V_2O_5 and Fe-Cr catalysts decreased substantially as depicted in Figures 1 and 2. Space velocity effects were much smaller with the Pt catalyst, as shown in Figure 3, in accord with results of Andersen et al (3-3).

Addition of up to 2000 ppm SO_2 into a test gas with typical flue gas (composition: 14% CO_2, 5% H_2O, 3% O_2, 1000 ppm NO) resulted in negligible change in the percent conversion of NO with 1100 ppm NH_3 on the V_2O_5 and Fe-Cr catalysts operating at 400°C. When SO_2 was added to the test gas with Pt catalyst at 250°C, however, a decrease in NO conversion occurred.

The results of the brief parametric tests in the TRW/UCLA project established selective reduction of NO with NH_3 as a promising means of abatement of NO and a preliminary economic analysis showed the process to be viable. Because of the lower operating temperature, platinum catalyst was attractive for use with low-sulfur fuels. The drawback of higher operating temperature with the non-noble metal catalysts was offset by their sulfur resistance.

The V_2O_5 and Fe-Cr catalysts, in view of their sulfur resistance and their tendency to promote reduction of NO to N_2 rather than N_2O as a final product, were of sufficient promise to merit additional investigation. A detailed parametric study to investigate more fully the effects of O_2, H_2O and CO_2 concentrations and to establish rate expressions for design of larger-scale reaction equipment was warranted. In addition, it was important to determine if the sulfur resistance shown over short-term exposure was maintained during extended periods of operation. These results of the TRW/UCLA project suggested further investigation of the non-noble metal catalysts which in the present project was designed to assess more fully the utility of V_2O_5 and Fe-Cr catalysts for use in the selective reduction of NO. As indicated in the Introduction, the primary objectives of the present program were to: optimize the compositions of the vanadia and iron-chromium catalysts for selective reduction of NO_x with NH_3, develop rate expressions for design use and perform long-term durability studies of optimum catalyst compositions in flue gas containing sulfur oxides.

A secondary objective of the project was to provide an update of current developments in NO_x catalysts in the published literature.

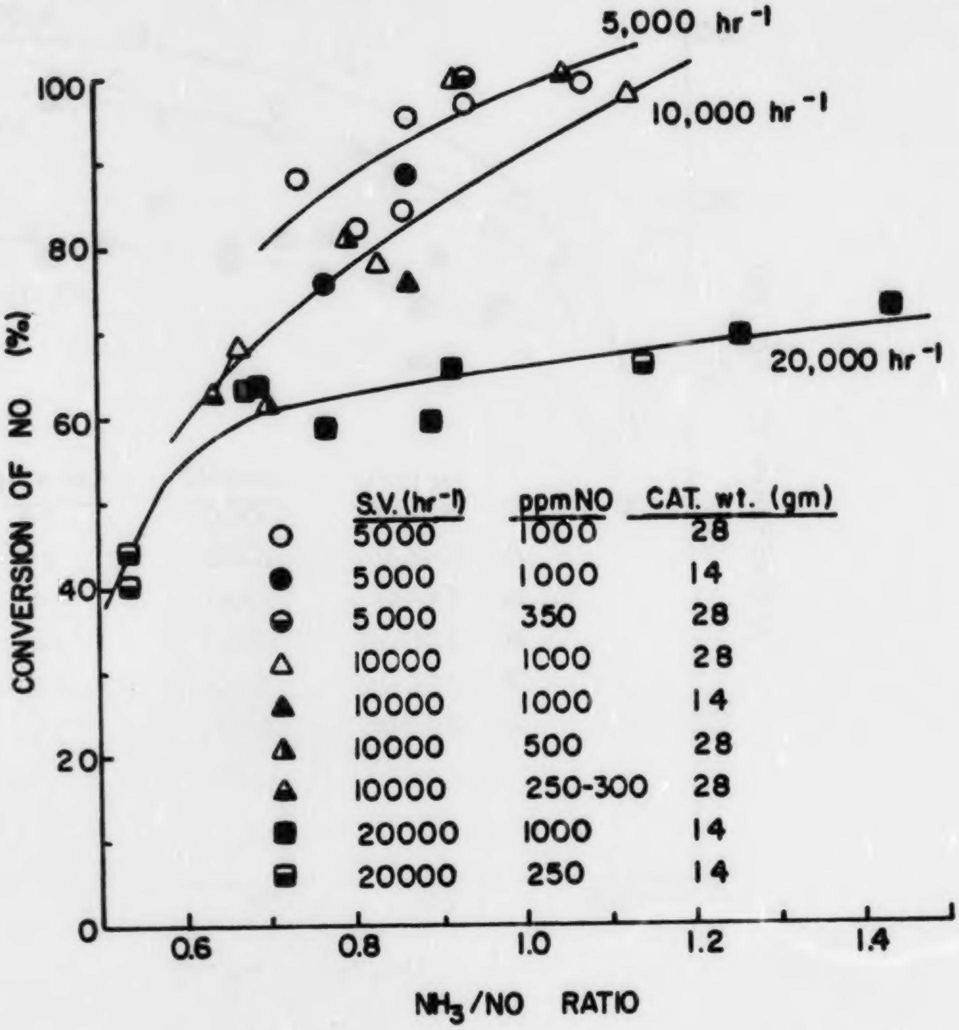

Figure 1. Reduction of NO with NH_3 on 10% V_2O_5 - Al_2O_3 (Harshaw V-0610T1/8) 14% CO_2, 5% H_2O, 3% O_2 present in N_2 carrier, 400°C.

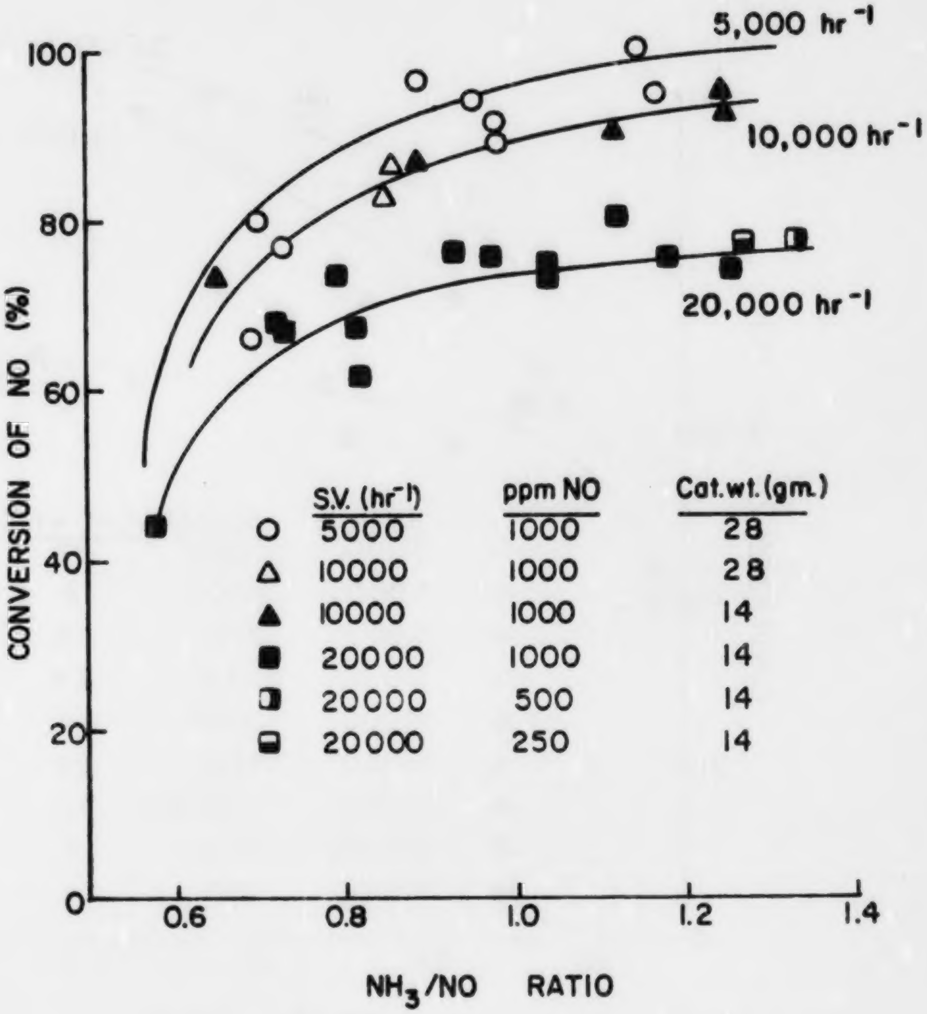

Figure 2. Reduction of NO with NH_3 on 10% Fe-Cr (5/1)-Al_2O_3 at 400°C. 14% CO_2, 5% H_2O, 3% O_2 in N_2 carrier.

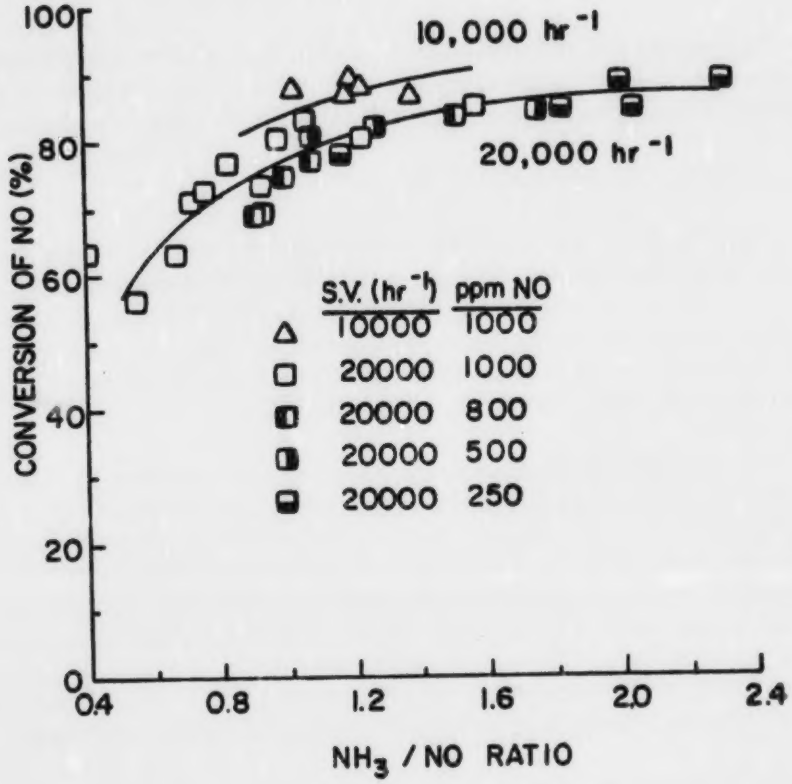

Figure 3. Reduction of NO with NH_3 on 0.5% Pt-Al_2O_3 (Engelhard) in simulated flue gas. 14% CO_2, 5% H_2O, 3% O_2 present in N_2 carrier; 250°C.

4. EXPERIMENTAL

The equipment and procedures used in the catalyst optimization studies and the parametric and durability tests are described in this section. A general description of the computer programs used for data reduction and correlation is also presented.

4.1. CATALYST PREPARATION

Vanadia and iron-chromium oxide catalysts were prepared by impregnation techniques using pre-formed alumina pellets as support material. Support materials tested in this project were (a) Filtrol Grade 86 cylinders, nominally 1/8 in. dia. x 3/16 in. length; (b) Alcoa H-151-8 spheres, 1/8 in. dia.; (c) Alcoa F-1-1/4-8 granules, irregularly shaped, up to about 1/4 in.; and (d) American Cyanamid Aeroban spheres (Al_2O_3-SiO_2 mixture), 3/16 - in dia.

The vanadia catalysts were prepared by impregnating dry carrier pellets with an aqueous solution of ammonium metavanadate and oxalic acid (2:1 molar ratio of NH_4VO_3 to $H_2C_2O_4 \cdot 2H_2O$). Because of solubility limitations a large amount of water was necessary for preparing catalysts with 10% or more V_2O_5 (final, dried form) preventing single-step solution absorption. The excess liquor was decanted from the pellets which were then partially dried. The remaining solution was absorbed by the semi-dried pellets. The pellets were fully dried at 160°C and then calcined in flowing air at 500°C to decompose the vanadate to V_2O_5 and burn off the organic acid. No other activation procedure was used before testing the catalysts.

Iron-chromium catalysts were prepared by impregnating alumina pellets with aqueous solutions of $Fe(NO_3)_3 \cdot 9\,H_2O$ and CrO_3. Complete absorption of the liquor was accomplished in a single step with the iron-chromium series of catalysts. The moist pellets were dried and calcined under conditions similar to those used with V_2O_5 catalyst. Complete details of the preparation of V_2O_5 and Fe-Cr catalyst showing highest activity are given in Section 5 (Results).

Table 1 summarizes the catalysts prepared and tested in this project. Also included in the table are several commercial catalysts tested for comparison purposes. In the vanadium series the percent of active material (V_2O_5) and type of carrier were of interest. With the Fe-Cr series the percent of active loading, as well as the Fe/Cr weight ratio was studied.

TABLE 1. LIST OF CATALYSTS TESTED

Prepared Catalyst	Carrier *
10% V_2O_5	Filtrol 86
15% V_2O_5	Filtrol 86
20% V_2O_5	Filtrol 86
25% V_2O_5	Filtrol 86
15% V_2O_5	Alcoa H-151-8
15% V_2O_5	Alcoa F-1-1/4-8
15% V_2O_5	Aeroban
5% Fe-Cr (1/1)	Filtrol 86
5% Fe-Cr (3/1)	Filtrol 86
5% Fe-Cr (9/1)	Filtrol 86
10% Fe-Cr (1/1)	Filtrol 86
10% Fe-Cr (3/1)	Filtrol 86
10% Fe-Cr (9/1)	Filtrol 86
10% Fe-Cr (9/1)	Aeroban
10% Fe-Cr (20/1)	Filtrol 86
10% Fe-Cr (50/1)	Filtrol 86
11% Fe-Cr (5/1)	Filtrol 86
20% Fe-Cr (1/1)	Filtrol 86
20% Fe-Cr (3/1)	Filtrol 86
20% Fe-Cr (9/1)	Filtrol 86
10% Fe_2O_3	Filtrol 86
20% Fe_2O_3	Filtrol 86

Commercial Catalysts

Vanadia

10% V_2O_5 Harshaw V-0601 T 1/8
(Al_2O_3 carrier)

Girdler Südchemie G101,
V_2O_5-K_2O-SiO_2 (V_2O_5/K_2O=1/2.7)

Fe-Cr

87% Fe-Cr (12/1) Girdler G-3A
(MgO, SiO_2 and graphite carrier).

10% Fe_2O_3 Harshaw Fe-0301-T1/8
(Al_2O_3 carrier)

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* alumina

4.2. BENCH-SCALE TEST APPARATUS

Optimization, parametric and durability studies were performed in a tubular, upflow reactor operating under isothermal conditions. Figure 4 shows, in schematic form, the reactor and associated equipment.

The reactor, as well as a tubular preheater, was maintained at preselected temperatures in an air-flow, fluidized sand bath heater using powdered alundum as the fluidized medium. A back pressure valve on the reactor outlet was adjusted to maintain a constant pressure in the system at all temperatures and to compensate for changes in pressure drop across the reactor bed when switching from inlet to outlet sampling modes.

Gaseous nitrogen, carbon dioxide and nitric oxide were mixed in a glass manifold after passing through needle-valves and rotameters. Nitric oxide flow was measured more accurately with a soap-bubble flowmeter.

Oxygen was added to the gas stream upstream of the vaporizer-mixer by appropriate injection of air. Ammonia and water were added at this point also. The ammonia was fed from an anhydrous liquid NH_3 tank using a rotameter for coarse flow adjustment. Water was injected in liquid form using a small rotary pump. The vaporizer-mixer was a 5/8 - in. dia. stainless steel tube, 10 inches long filled with 1/8-in. glass beads. The tube was lagged with a heating tape and insulated to maintain a temperature of at least 350°C.

When SO_2 was used, its injection point was immediately upstream of the vaporizer-heater. Earlier attempts to introduce SO_2 at the common entry for NH_3, air and H_2O invariably led to plugging of the system due to salt formation at low temperatures.

The reactor was a stainless steel tube with end fittings containing stainless steel screen discs to retain the catalyst. Two reactor sizes were employed. The standard size used in most of the tests was constructed of 5/8 in. dia. tubing to contain 14 ml. of catalyst. The second reactor was constructed of 1.0 in. tubing and had an internal volume of 28 ml.

4.3. ANALYTICAL EQUIPMENT AND PROCEDURE

Inlet and outlet analyses for NO were made with a Beckman Model 315A non-dispersive infrared analyzer. Routine analyses for NO_2 were made with a Beckman Model 77 visible-light analyzer.

Nitrous oxide (N_2O) concentrations were determined with a Perkin-Elmer Model 990 chromatograph with a 6 ft, 1/8 in.-dia. column of Porapak Q operating at room temperature. The temperature was varied periodically up to 150°C during analyses of outlet gases in the presence of SO_2 to check for the production of H_2S and COS. In view of the overall oxidizing nature of the test atmosphere, the presence of either

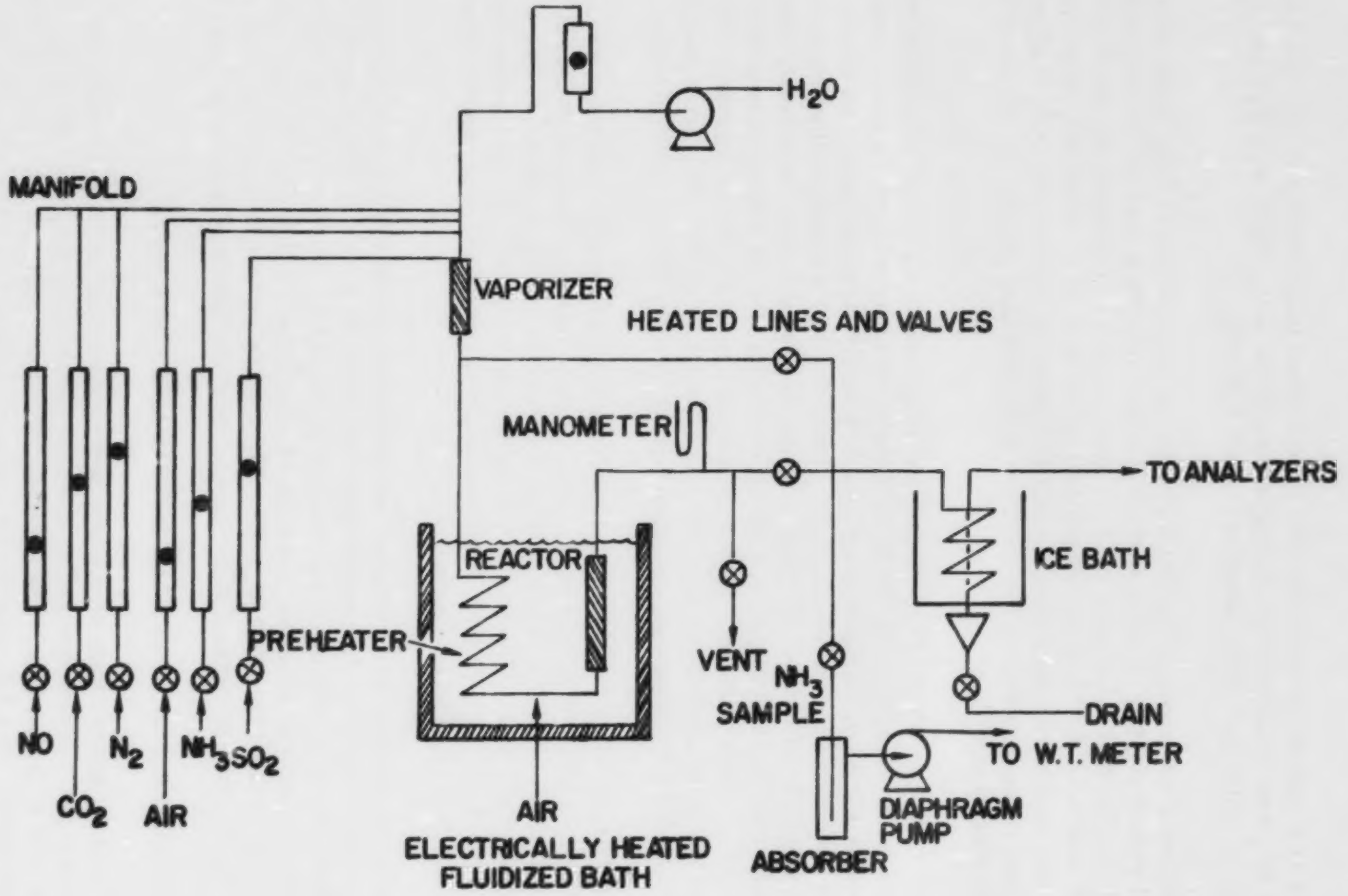
H$_2$O
MANIFOLD
VAPORIZER
HEATED LINES AND VALVES
MANOMETER
TO ANALYZERS
REACTOR
ICE BATH
PREHEATER
VENT
NH$_3$
SAMPLE
DRAIN
TO W.T. METER
NO
N$_2$
NH$_3$
SO$_2$
CO$_2$
AIR
AIR
ELECTRICALLY HEATED
FLUIDIZED BATH
ABSORBER
DIAPHRAGM
PUMP

Figure 4. Catalyst test apparatus.

species was considered unlikely; neither was found in any instance (surface dissociation of NH_3 was considered to be a possible source of H_2 which with H_2O and CO_2, through the water gas shift, could be a source of CO).

Ammonia was analyzed by two methods. In the absence of SO_2, a wet titration method was employed. A volume of gas, measured with a wet-test meter, was drawn through an absorber containing 4% boric acid in water. The absorber contents were titrated with 0.04 N HCl using bromcresol green indicator. When SO_2 is present, sulfite and sulfate interfere with the HCl titration. In these tests, NH_3 analyses were performed on the contents of the absorber with an Orion specific ion electrode and a Keithley electrometer.

Sulfur dioxide was determined indirectly by measuring separately sulfur trioxide and total oxides of sulfur. In the former measurement, a known volume of gas was absorbed in a train of bubblers containing 80% isopropanol in water. An aliquot of the solution was titrated with 0.01 N $BaClO_4$ using Thorin indicator. The precipitate formed remains colloidal in the high concentration of non-aqueous solvent. Thorin, which adsorbs on the precipitate, undergoes a color change from yellow to pink in excess Ba^{++}. Sulfur dioxide, carbon dioxide and ammonia do not interfere with the titration which is specific for $SO_4^{=}$ (4-1). Oxidation of sulfite ($SO_3^{=}$ from dissolved SO_2) is avoided by maintaining the absorbent at ice temperature until titration.

Total oxides of sulfur were determined by passing a known volume of gas through bubblers containing basic 1.5% H_2O_2 (equi-volume mixture of 0.2 N NaOH and 3% H_2O_2). Any SO_2 absorbed in the bubbler was oxidized by H_2O_2 to $SO_4^{=}$. The contents of the bubbler were acidified to prevent formation of $Ba(OH)_2$ in the ensuing titration, boiled to drive off CO_2, passed through a cation exchange resin to remove interfering sodium and then titrated as above.

The sulfur dioxide concentration was calculated as the difference between total oxides of sulfur and SO_3. Although there was some difficulty in observing the end point with laboratory-prepared solutions of H_2SO_4 as a source of $SO_4^{=}$, it was found that the technique was accurate to within 3% when gases with known concentrations of SO_2 were absorbed in basic H_2O_2. The difficulty in applying the method to the solutions of H_2SO_4 may be related to the dibasic acid equilibrium between $SO_4^{=}$ and HSO_4^{-}.

4.4. STANDARD FLUE GAS COMPOSITION AND FLOW RATE

In Task 1, optimization of catalyst composition was based on performance using a single test gas mixture that had as the sole variable the NH_3 concentration. This mixture, called in this report the "standard flue gas" was also used in most of the durability tests. The NH_3 concentrations were either near-stoichiometric for reduction of NO to N_2 or in excess.

The standard flue gas contained 12% CO_2, 5% H_2O, 3% O_2 and 1000 ppm (parts per million by volume) NO in N_2. Ammonia was either 650-750 ppm (670 ppm is stoichiometric) or 1000-1100 ppm. By using the 14 ml. reactor, the normal total gas flow rate was 280 liters/hr (STP: 0°C, 1 atm) providing a space velocity of 20,000 hr^{-1}. The 28 ml. reactor at the normal flow rate provided a space velocity of 10,000 hr^{-1}.

4.5. TEST PROCEDURE

The typical start up procedure was to adjust N_2 flow rate temporarily to 266 liters/hr (corresponding to total dry gas in the flue gas mixture) and to introduce NO at 1000 ppm in order to span the NO analyzer. The N_2 flow rate was then decreased to correspond to its concentration in the flue gas (allowing for the N_2 introduced in the air stream) and the other gas species (except for NH_3) were added.

The sand bath and the vaporizer/mixer heaters and those on the lines to the absorbers were set to appropriate power levels. Using the analyzer calibration curves, the desired NO concentration was obtained by adjusting the feed needle valve. The desired ammonia concentration was estimated with a rotameter. The gas mixing process was allowed to stabilize for about 30 minutes and the inlet stream was sampled for NH_3, using the titration method described above, and the NH_3 concentration adjusted as required.

If SO_2 was to be added, it was introduced using a rotameter as a guide. Analysis of the inlet stream followed the described procedure. After introduction of SO_2, all subsequent inlet and outlet NH_3 analyses were performed with the Orion probe. Outlet and inlet analyses were routinely performed using as a criterion for steady-state the constant, analyzed concentrations of NO, N_2O, NO_2, and NH_3 for at least 30 minutes.

In the parametric studies (Task 2), NO, NH_3, O_2, CO_2 and H_2O concentrations, as well as temperature and flow rate, were all varied to provide large perturbations from the standard conditions.

In the durability studies with SO_2 present, periods of steady-state operation under SO_2-free conditions were frequently followed to provide comparison data and to check for catalyst degradation.

4.6. DATA REDUCTION

Development of empirical rate expressions proceeded according to the following sequence. A rate expression of the following form was assumed:

$$r, \text{rate} = k\, p_{NO}^{n}\, p_{NH_3}^{m}\, p_{O_2}^{q} = Ae^{-E/RT}\, p_{NO}^{n}\, p_{NH_3}^{m}\, p_{O_2}^{q}\ \frac{\text{g-moles NO}}{\text{hr, gm catalyst}} \qquad (1)$$

A computer program was employed that determines an experimental rate constant (k_{exp}) for each data point and then determines by linear regression the best-fit A and

E values for all data points. Specifically, the computer solves the combination of (1) and the plug flow mass balance,

$$\frac{W}{F} = \int_0^x \frac{dx}{r} \, , \tag{2}$$

to give

$$\frac{k_{exp} W (p^o_{O_2})^q}{Q \rho p^o_{NO} \pi} = \int_0^{x_{exp}} \frac{dx}{p^{o\,n}_{NO} (1-x)^n p^{o\,m}_{NH_3} \left(1 - \frac{x}{S}\right)^m} \tag{3}$$

where F = inlet feed rate of NO, gm mole NO/hr

W = weight of catalyst, gm.

Q = volumetric flow rate, ℓ/hr.

ρ = molar density of feed, gm mole/ℓ

$p^o_{NO}, p^o_{NH_3}, p^o_{O_2}$ = inlet partial pressures, atm.

$p_{NO}, p_{NH_3}, p_{O_2}$ = instantaneous partial pressures, atm (Equation 1)

x = conversion of NO

= $(p^o_{NO} - p_{NO})/p^o_{NO}$

n, m, q = reaction orders

k_{exp} = experimental rate constant, $\frac{\text{gm moles}}{\text{hr-gm catalyst-(atm)}^{n+m}}$

S = stoichiometric factor = 3/2

π = reactor pressure, atm.

The computer calculation determines the k_{exp} for a large number of combinations of n, m and q. For each combination, linear regression of k_{exp} as a function of (1/T) is performed, giving the constants A and E (see Eq. 1). The program then determines the sum of squares (σ) of the difference between specific rate constants calculated from the regression parameters and the experimental values, k_{exp},

$$\sigma = \sum (k - k_{exp})^2 \tag{4}$$

The minimum sum of squares, found by inspection of the output record, corresponds to the combination of n, m and q which best describes the data.

The resulting rate expression describes global kinetics in the test system employed. This rate expression, by itself, cannot be used to predict performance in other reactors unless it can be demonstrated that in the generation of experimental data, there were no bulk diffusion and pore diffusion effects. That is, for design purposes, an intrinsic (surface) rate expression must be available from which global rates in other equipment can be predicted.

To determine intrinsic rate expressions a second computer program was employed. This program determines, in a step-by-step manner from reactor inlet to outlet, the surface concentrations of reactants and surface temperatures based on mass and heat transfer correlations, and an effectiveness factor, η (defined below), which parameterizes internal diffusion effects. By assuming an intrinsic (surface) rate expression, the degree of conversion of NO is calculated throughout the catalyst bed. The rate at any point (i) is determined by

$$r_i = \eta k_s p_{NO}^{n} p_{NH_3}^{m} p_{O_2}^{q} \qquad (5)$$

where k_s is the intrinsic rate constant and the partial pressures are those at the outer surface, as determined in the bulk diffusion effect calculations. Calculated conversion at the reactor outlet is compared with the experimental value and the process is repeated with modified forms of the assumed intrinsic rate expression until agreement is obtained. Effectiveness factors were determined for each reactor segment in a step-wise procedure using relationships involving the Thiele diffusion modulus (4-2, 4-3, 4-4). Near-unity values of η indicate that pore diffusion effects are negligible. For convenience, the calculation procedure begins by assuming that the intrinsic rate parameters are those determined from the correlation of the global rates as described earlier (i.e., initially assuming negligible mass transfer resistances).

The final form of the intrinsic rate expression can be utilized for the determination of global rates of reaction at any scale of operation (assuming catalyst surface activity is unchanged) by calculating surface concentrations and temperatures and effectiveness factors for the desired conditions. More details of the computer program have appeared in other publications (4-4, 4-5).

4.7. LITERATURE ACQUISITION (Task 3)

The computerized literature survey at the UCLA library, augmented by compilations from APTIC, provided appropriate references of articles and patents pertinent to NO_x catalysis. A thorough analysis of each reference was not attempted in the project. This survey was used to identify particularly promising catalysts used for the selective reduction of NO with NH_3 and to obtain, where possible, pertinent operating data.

The primary source of literature was from patent files and, of these, by far, the predominant sources were Japanese. A number of these patents were translated. The majority of the literature sources could not be obtained during the course of the project. Reliance on abstracts as published by the standard literature-retrieval abstracting services was made to provide some information. A list of references was made, including such pertinent data as was available, and is presented in Appendix A.

5. RESULTS

The results of the experimental effort in each of the three tasks are summarized in the following sections. Tasks 1 and 2 were performed chronologically, as described, while Task 3 was performed as a continuing effort throughout the program.

5.1. Task 1. OPTIMIZATION OF COMPOSITION AND DEVELOPMENT OF CHEMICAL SPECIFICATIONS OF VANADIA AND IRON-CHROMIUM CATALYSTS

Several catalysts were prepared with variations in the amount of active material and, in the case of Fe-Cr catalysts, in the ratio of the metal constituents. In addition, several commercial catalysts were obtained and tested. Tests in simulated flue gas containing 12% CO_2, 3% O_2 and 5% H_2O and either stoichiometric or excess NH_3 were performed to assess the effects of catalyst composition on catalyst activity. Temperature was the only variable (other than the two NH_3 concentrations) in the screening studies. Although some of the catalysts, which have been shown to be inferior under the present study, may be more active under other conditions, such operating modes will probably be outside of practical operating regimes.

Prior to conducting the screening tests a check was made to determine if reaction occurred in the empty reactor and feed systems and to determine the extent of conversion of NO to NO_2 in colder parts of the system. Negligible conversion of NO to N_2 or N_2O was observed, although a small amount of NO_2 was formed, depending on O_2 concentration. In general, NO_2 production was less than 100 ppm with 1000 ppm NO in 3% O_2. It was deduced that the NO_2 was formed in the NO_2 analyzer itself which has a 15 minute residence time at the sampling flow rate employed. Inlet and outlet concentrations of NO_x as tabulated in this report have been corrected to include the small NO_2 concentrations observed.

5.1.1. Vanadia Catalysts

Several vanadia-alumina catalysts were prepared with various concentrations of V_2O_5. Compositions are listed in Table 1. Filtrol Grade 86 Al_2O_3 (1/8-in. dia. x 3/16 - in. long cylinders) was used as support material in the first series of preparations. Catalysts containing 10, 15, 20 and 25% (by weight) V_2O_5 were prepared by impregnating the Al_2O_3 pellets with aqueous solutions of ammonium metavanadate (oxalic acid was used to increase the solubility of the vanadate). The impregnated pellets were dried and then calcined in air at 450°C to form the vanadium oxide and drive off ammonia and organics.

Catalyst activity was determined in a simulated flue gas stream containing 12% CO_2, 5% H_2O, 3% O_2 and 1000 ppm NO with approximately 700 ppm or 1100 ppm NH_3 (balance N_2). The 700 ppm NH_3 concentration represents a near-stoichiometric amount for conversion of the NO to N_2. The 1100 ppm NH_3 represents a 65% excess of NH_3. Temperature was varied from 100 to about 450°C with each catalyst. Flow rates were maintained at 280 liters/hr (STP) to provide a space velocity of 20,000 hr^{-1} (STP).

Figures 5 and 6 show conversion of NO as a function of temperature for 700 and 1100 ppm NH_3, respectively. Also included in the figures are data for the commercial 10% V_2O_5-Al_2O_3 catalyst (Harshaw V-0601T 1/8) obtained in the previous TRW/UCLA project. Figure 5 shows that the prepared V_2O_5 catalysts were of approximately equivalent activity at the stoichiometric NH_3/NO ratio. Between 300-450°C the prepared catalysts were more active than the commercial catalyst.

With excess NH_3 (Figure 6) highest conversion levels for temperatures above 250°C were obtained with the 15% catalyst. However, the differences in activity of the prepared catalyst were not large. The commercial V_2O_5 catalyst was as active as the 10 and 25% catalysts at the higher temperatures.

Figure 7 shows that, at 300 and 400°C, the optimum composition occurs at about 15% V_2O_5. The decreased activity of the 25% catalyst may be due to excessive loading of V_2O_5 in the catalyst pores with a corresponding decrease in surface area.

The surface area of the Harshaw catalyst was reported as 115 sq. m/gm. Using a chromatographic surface area measurement technique, the surface area was determined to be 103 sq. m/gm, indicating good agreement with the reported Harshaw BET value. Surface areas of the 15 and 25% V_2O_5 catalysts were 198 and 154 sq. m/gm, respectively, lending support to the hyposthesis that reduced activity above 15% V_2O_5 is related to decreased surface area.

Two additional catalysts were prepared using Alcoa activated aluminas as support material for comparison with catalysts using the Filtrol-alumina support. The optimum level of 15% V_2O_5, as determined in the studies with the Filtrol support, was impregnated on Alcoa F-1-1/4-8 and H-151 aluminas. The former was in the form of irregular granules up to 1/4 in. Particles of about 1/8-in. were selected for use. The H-151-8 alumina was in the form of 1/8-in spheres.

The F-1-1/4-8 and H-151 catalysts were tested in simulated flue gas with 1000 ppm NO and 1100 ppm NH_3. Figure 8 shows that the F-1-1/4-8 support provided significantly higher activity than H-151. Also shown in the figure are data for Filtrol Grade 86 catalyst containing 15% V_2O_5 for runs with 1100 ppm NH_3. Above 380°C the Filtrol and the F-1-1/4-8 supported catalysts were equivalent in activity. Below that temperature the latter support appeared distinctly superior. In particular, maximum conversion (about 95%) occurred at only 340°C compared to a slightly lower conversion maximum at 390°C with Filtrol alumina.

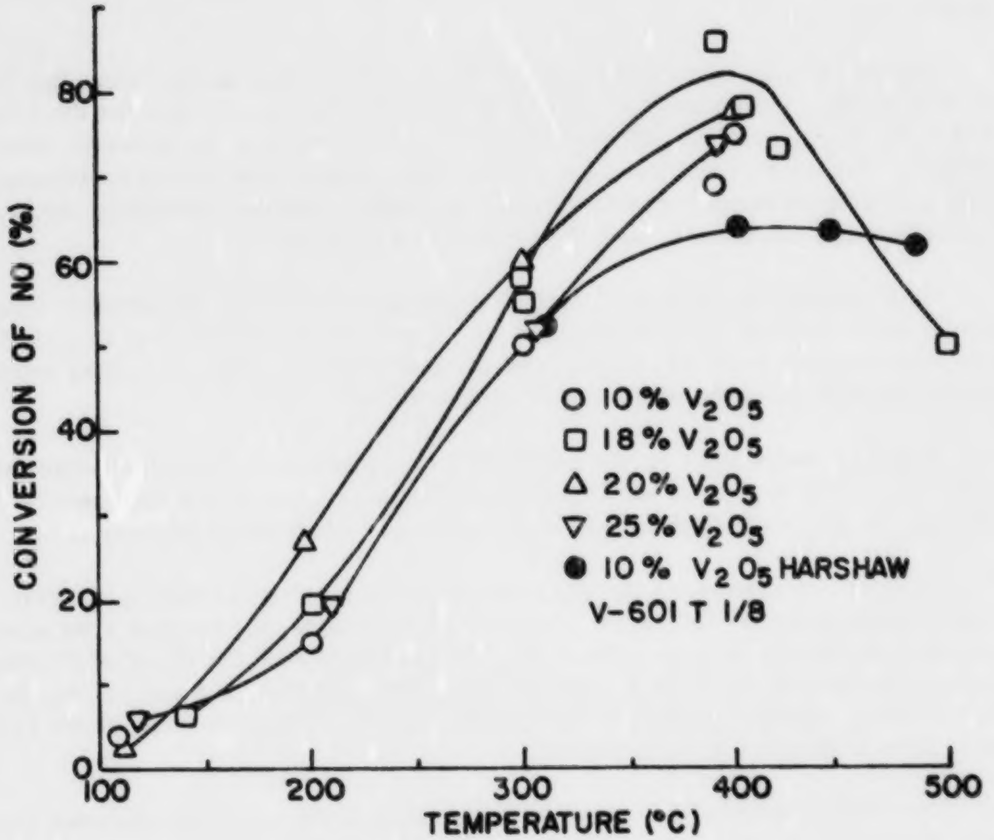

Figure 5. Reduction of NO (1000 ppm) with NH_3 (700 ppm) on V_2O_5-Al_2O_3 catalysts in simulated flue gas. (s.v. = 20,000 hr^{-1}).

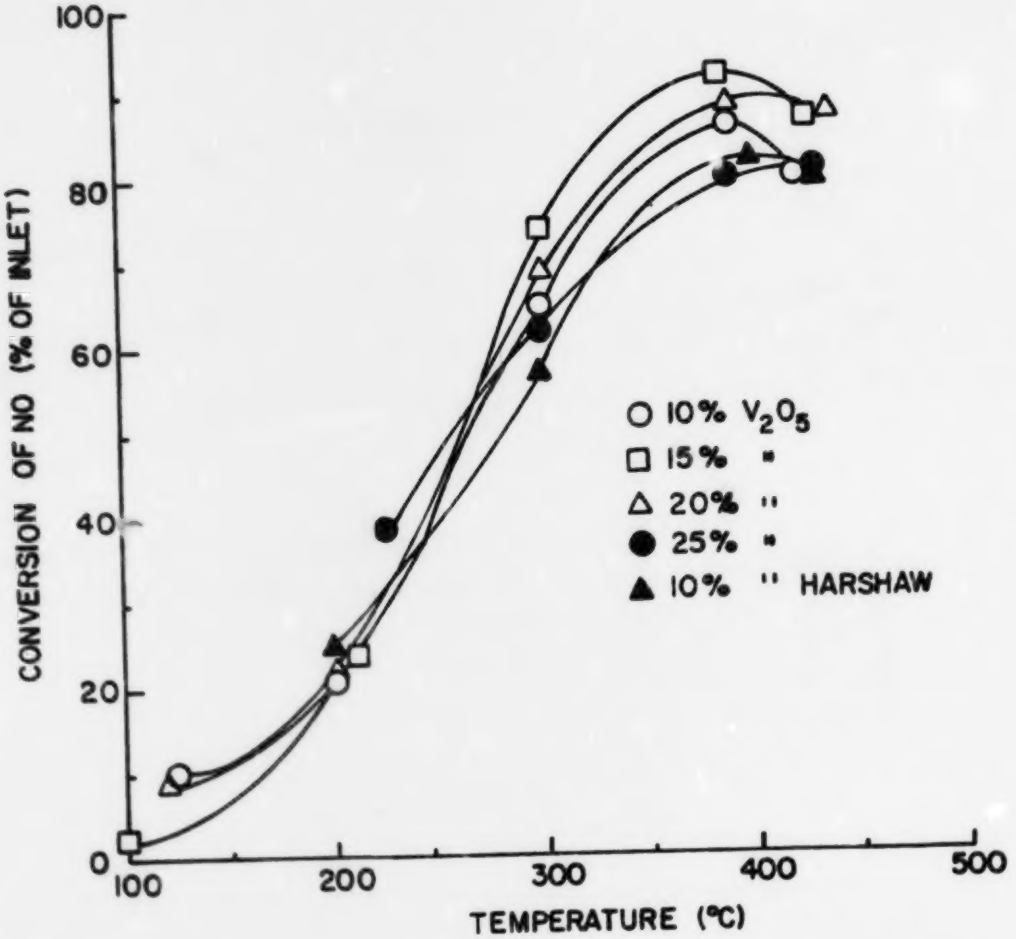

Figure 6. Reduction of NO (1000 ppm) with NH_3 (1100 ppm) on $V_2O_5-Al_2O_3$ catalysts in simulated flue gas. (s. v.=20,000 hr^{-1}).

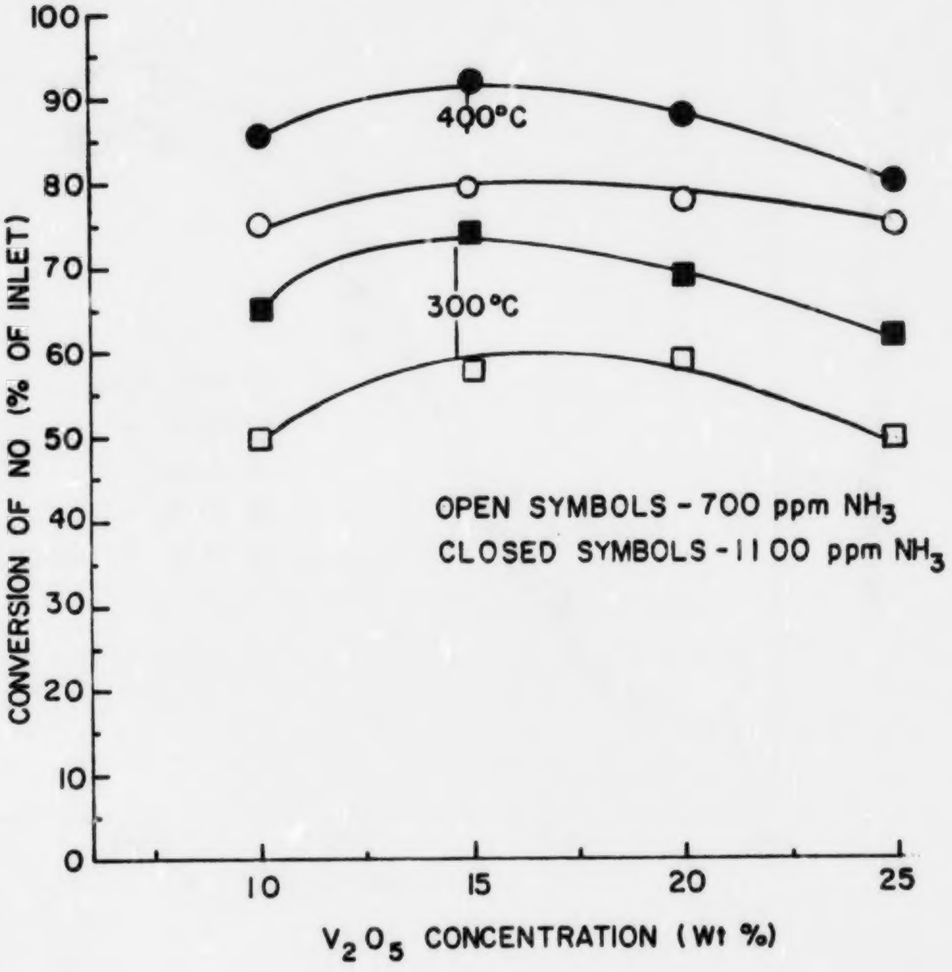

Figure 7. Influence of V_2O_5 concentration on catalyst activity in simulated flue gas. (s.v.= 20,000 hr^{-1}).

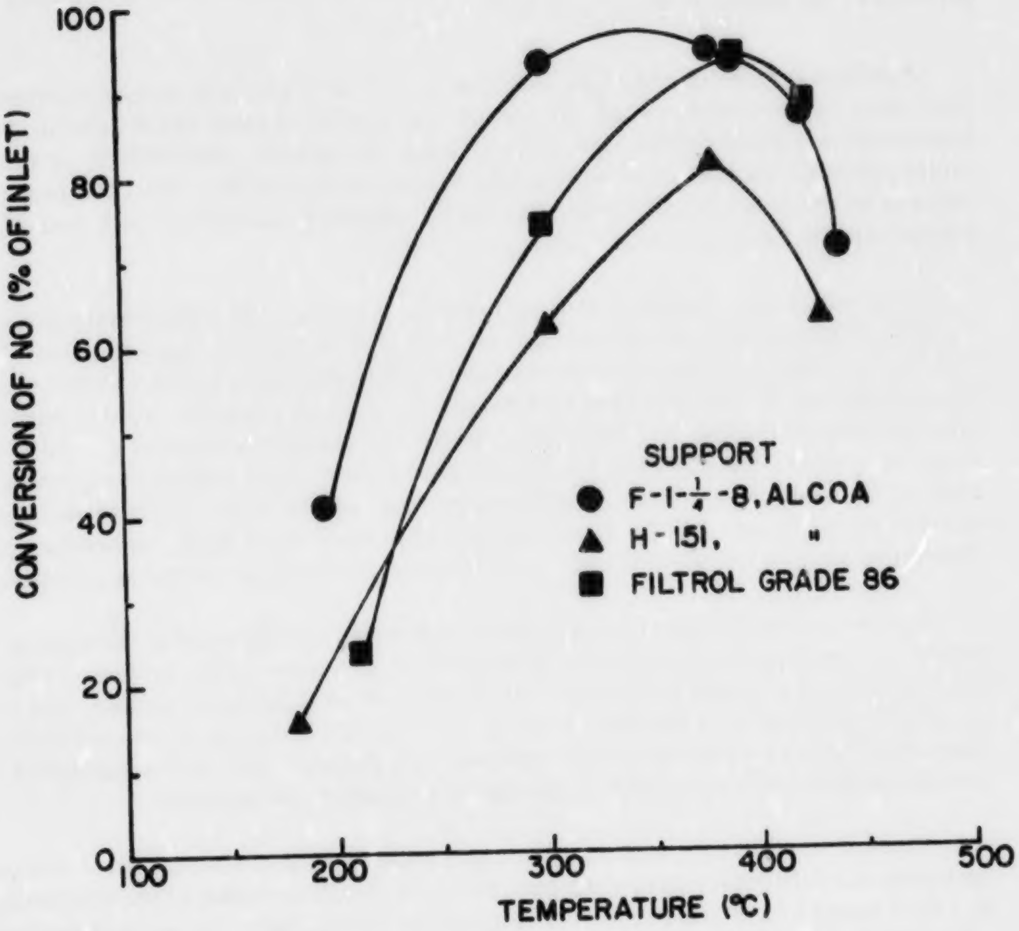

Figure 8. Effect of Al_2O_3 support on NO conversion in simulated flue gas, with 15 % V_2O_5 catalysts. 1000 ppm NO, 1100 ppm NH_3. (s.v.= 20,000 hr^{-1}).

With all the V_2O_5 catalysts tested, no N_2O was detected in the exhaust gas stream. NH_3 consumption was, in general, less than 65% in excess of the theoretical amount required to achieve total reduction of NO to N_2. Up to the temperature of maximum NO conversion with each catalyst, above the maximum, NH_3 usage increased sharply and, at about 420°C, reached values more than double that required for the reduction of NO. It is probable that some of the NH_3 which reacted at higher temperatures was oxidized directly to NO.

Additional tests of 15% V_2O_5 catalyst on Alcoa F-1-1/4-8 activated alumina carrier were performed to verify the initial, promising results which indicated somewhat improved performance over the Filtrol Grade 86 carrier, particularly at the lower temperatures. Repeat runs were made in simulated flue gas with 1100 ppm NH_3 to determine the repeatability of earlier data. Figure 9 shows that both sets of data are within expected experimental error.

For reference, Figure 9 also includes data for the 15% , laboratory-prepared catalyst on Filtrol Grade 86 carrier and the two sets of data for the commercial Harshaw 10% V_2O_5 catalyst. The first set was obtained in the earlier UCLA/TRW program. The second set of data was obtained during the present project. Again, satisfactory repeatability is indicated. The data indicate that the Alcoa support provides superior activity to either the commercial Harshaw or Filtrol Al_2O_3-supported, prepared catalyst at lower temperatures. On face value the results would seem to indicate better carrier performance for the Alcoa compared to the Filtrol type. However, other characteristics were considered in the selection of the most suitable carrier.

It was noticed during testing of the Alcoa catalyst that considerable fines were produced, leading in one test to excessive back pressure in the system. This particular activated alumina is not normally marketed as a catalyst carrier, but is usually used as a dessicant. Even though Alcoa reports that F-series activated aluminas have high abrasion and water-breakage resistances, the present results suggested a comparison of the crush strengths of the various catalysts of interest.

Table 2 summarizes results of compression testing of the Harshaw commercial catalyst, the laboratory prepared catalysts and the Filtrol and Alcoa carriers. The H-151-8 is spherical and F-1-1/4-8 is granular; thus, only one mode of testing is given for these materials. The Alcoa aluminas are noticibly weaker than either the Filtrol or Harshaw catalysts. The low strength of the Alcoa aluminas eliminated them from consideration for NO_x control. Also, the degree of powdering observed for this small, bench scale of operation is expected to be magnified under full-scale operation.

It should be mentioned that another commercial V_2O_5 (10%) catalyst prepared by Filtrol was tested in the earlier project and was found to be physically inferior to the Harshaw catalyst. More importantly, it was found to be chemically unstable. It is interesting that only the Filtrol Grade 86 carrier is reasonably strong. No information is available to indicate if the Filtrol V_2O_5 catalyst contains the Grade 86 Al_2O_3.

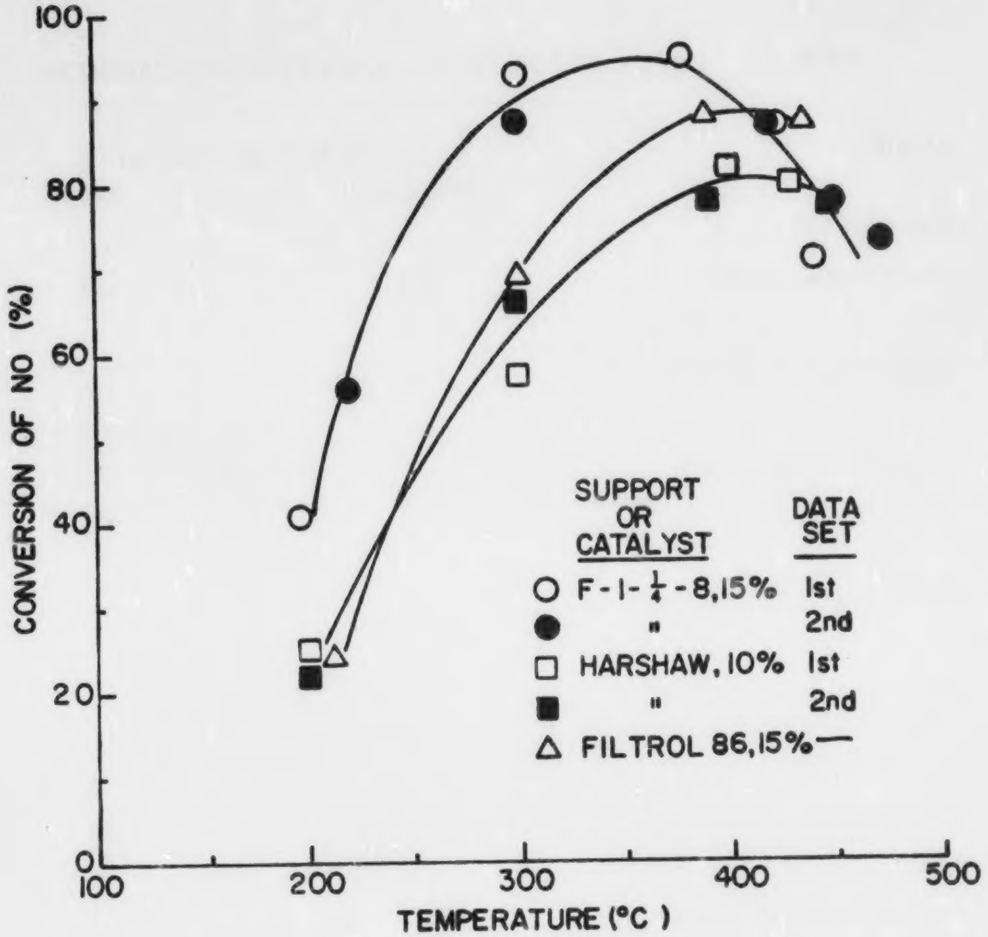

Figure 9. Reduction of NO (1100 ppm) with NH_3 (approx. 1100 ppm) on prepared and commercial V_2O_5 catalysts in simulated flue gas. (s.v.=20,000 hr^{-1})

TABLE 2. CRUSH STRENGTH OF CATALYSTS AND CARRIERS

Material	Crush Strength * (lb force) Longitudinal	Radial
Harshaw 10% V_2O_5 commercial catalyst	17	19
Alcoa H-151-8 carrier	9	--
Alcoa H-151-8 carrier with 15% V_2O_5	5	--
Alcoa F-1-1/4-8 carrier	7	--
Alcoa F-1-1/4-8 carrier with 15% V_2O_5	5	--
Filtrol Grade 86 carrier	20	12
Filtrol Grade 86 carrier with 15% V_2O_5	24	13.5

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* All data are averages of 4 tests.

A brief series of tests was performed with catalysts using Aeroban ZW-1470 carrier (Al_2O_3 - SiO_2) from American Cyanamid to compare activity with catalysts using the Filtrol Al_2O_3. Figure 10 shows that for a feed gas containing 1000 ppm NO and excess NH_3, there is substantially no difference between the two catalysts. With stoichiometric NH_3 the Filtrol-supported catalyst appears slightly superior.

A commercial vanadia catalyst specifically developed for SO_2 oxidation in contact sulfuric acid plants was tested to complete the survey of in-house and commercially prepared V_2O_5 catalysts. The commercial catalyst was Girdler-Südchemie type G-101. This catalyst was particularly attractive because of its very high physical strength (37 lb.), as reported by the manufacturer. The actual V_2O_5 content of the catalyst is unknown. Information provided by the manufacturer indicate a silica carrier impregnated with vanadium and potassium salts, the latter in a ratio (as K_2O) with V_2O_5 of about 2.7/1. Although previous work in this laboratory has shown that even small concentrations of alkali metals in V_2O_5 catalysts reduced the catalytic activity for reduction of NO with NH_3, it was of interest to determine the activity of this commercial catalyst containing K_2O.

Figure 11 shows that the G-101 catalyst has practically no activity for NO conversion in simulated flue gas containing 12% CO_2, 5% H_2O and 3% O_2. Above 380°C, effluent gas analysis, in fact, showed NH_3 oxidation with O_2 to NO and N_2. The present results combined with the previous observation of alkali metal poisoning indicate that commercial sulfuric acid catalysts containing alkali metal promoters will probably not be effective for NO_x reduction with NH_3. Therefore, for the present, simultaneous reduction of NO with NH_3 and oxidation of SO_2 to SO_3 does not appear promising, if the alkali metal additives are required for the latter reaction.

The combined results of the screening studies and the crush strength measurements indicate that the 15% V_2O_5 - Filtrol Grade 86 is the most promising catalyst tested. Therefore, this catalyst was selected for further study in both the parametric test program (Task 2) and the SO_2 durability studies.

5.1.2. Iron-Chromium Catalysts

In addition to the optimum amount of active material (total iron and chromium oxides) deposited on the carrier it was necessary to determine the optimum ratio of iron to chromium with this series of catalysts. Previous experience at this laboratory indicated the maximum total oxide loading that could be attained in a single-step impregnation of the carrier was about 20%. Initially, a matrix which involved tests of a series of catalysts of variable oxide loading and Fe:Cr ratio was designed to include concentrations up to 20% oxides and variation of the Fe:Cr ratio about the value used by Schmidt and Schulze (3-2) (i.e., about 5/1 Fe:Cr).

Table 3 lists the compositions of nine catalysts in the initial survey.

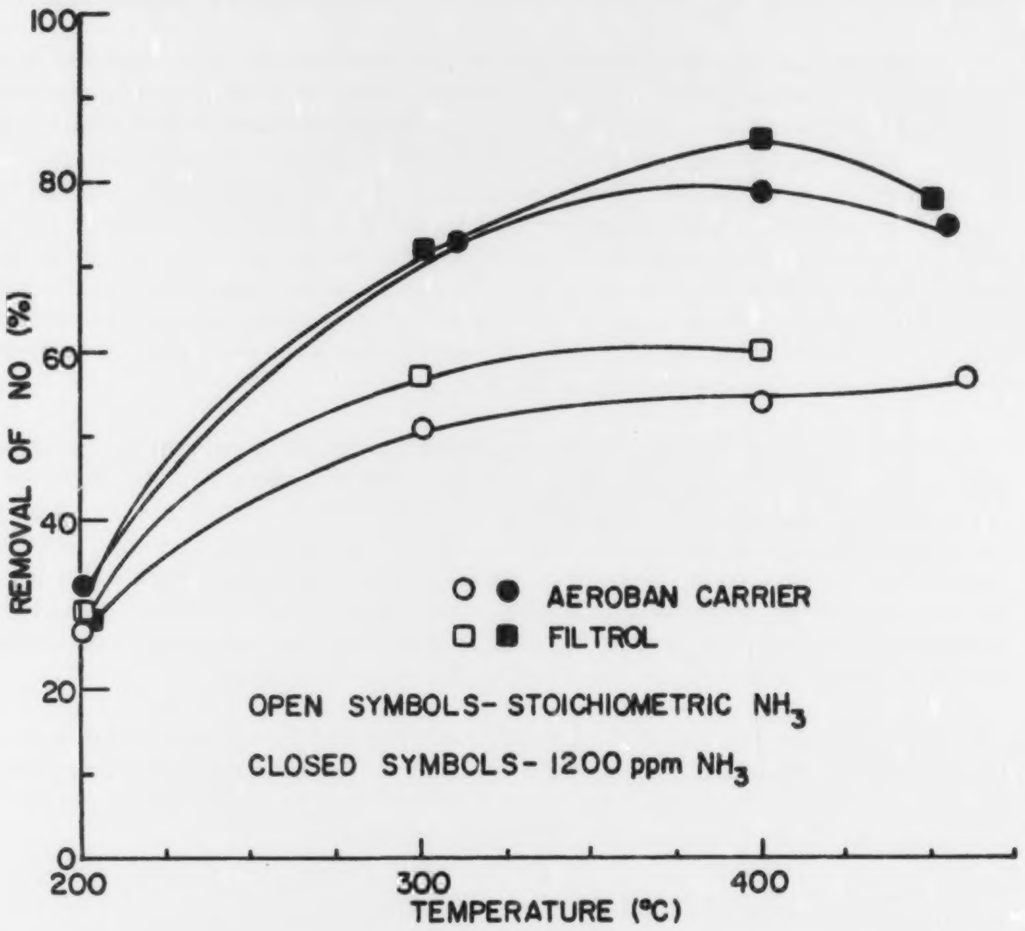

Figure 10. Removal of 1000 ppm NO with NH_3 in simulated flue gas on 15% V_2O_5 catalysts. 12% CO_2, 3% O_2, 5% H_2O, balance N_2. (s.v.=20,000 hr^{-1}).

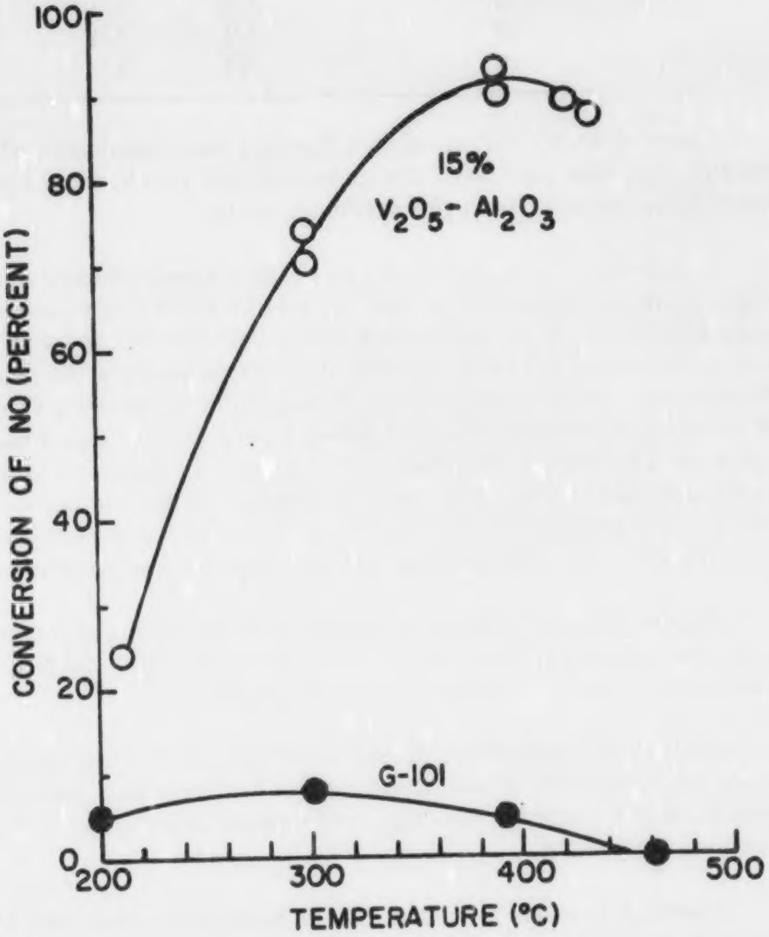

Figure 11. Comparison of 15% V_2O_5- Al_2O_3 and commercial V_2O_5- K_2O - SiO_2 catalysts. 1000 ppm NO and 1100 ppm NH_3 in simulated flue gas. (s.v.= 20,000 hr^{-1}).

TABLE 3. IRON-CHROMIUM TEST MATRIX

Percent Active Material (wt. % oxides)	Iron / Chromium Ratio (weight basis)		
5	1/1	3/1	9/1
10	1/1	3/1	9/1
20	1/1	3/1	9/1

Tests of activity in simulated flue gas were conducted with either 700 or 1100 ppm NH_3 and 1000 ppm NO. Temperature was varied from 200 to 460°C. Table 4 summarizes the results of the screening tests.

In general, the catalysts showed either maximum activity at about 400°C, or a broad maximum from 300° to 400° C, except for the 20% Fe-Cr oxide catalysts. Large amounts of N_2O, which maximized between 385 and 450°C (i.e., at temperatures in excess of the point of maximum conversion), were produced on these three 20% catalysts. At the temperature of maximum conversion (300°C) the activity of the 20% catalysts increased with increasing iron content. This observation is shown clearly (see open points for the 20% active material) in Figures 12 and 13. At 400°C (closed points) and excess NH_3, Figure 12 shows that maximum conversion occurs at 10% loading. The catalytic activity of the 9/1 catalyst is superior to both the 1/1 and the 3/1 catalysts at the 10% loading with the latter two of approximately equivalent activity.

Figure 13 shows a trend of higher activity with increasing iron levels at 400°C and a near-stoichiometric NH_3 concentration at both 5 and 20% loading. All 10% catalysts were of approximately equivalent activity.

Analysis of these initial results indicates that optimum catalyst composition for excess NH_3 operation at 400°C appears to be 10% active material (Fig. 12). At 300°C, however, a 20% catalyst with high iron content is indicated for use with either excess or stoichiometric NH_3.

Previous results (1-2, see Fig. 2) have shown that with 11% Fe-Cr (5/1) catalysts operating at stoichiometric NH_3, conversion of NO at 400°C can be increased from about 60% to about 80% by decreasing the space velocity from 20,000 hr^{-1} to 5000 hr^{-1}. With excess NH_3, however, the improvement in conversion by operation at lower space velocities is more substantial. For purposes of catalyst selection for parametric studies it was assumed that a minimum space velocity of 10,000 hr^{-1} would be acceptable for practical reactor sizes.

Excess usage of NH_3 does reflect strongly, of course, on the operating cost. A

TABLE 4. RESULTS OF ACTIVITY TESTS WITH Fe-Cr CATALYSTS

Catalyst Active Material	Fe-Cr	Temp. (°C)	Inlet Gas		Outlet Gas			NO Conversion (%)
			NO (ppm)	NH_3 (ppm)	NO (ppm)	NH_3 (ppm)	N_2O (ppm)	
5%	1/1	205	940	1036	880	942	0	6
		300	940	1036	610	600	0	35
		400	940	1036	315	94	50	66
		465	940	1036	450	0	50	52
5%	1/1	200	980	775	905	730	0	8
		300	980	775	683	345	0	35
		400	980	730	510	0	0	48
		478	984	775	515	0	0	48
10%	1/1	200	960	1100	805	1012	0	16
		295	960	1100	430	412	0	55
		400	1040	1140	325	0	0	68
		450	930	1100	530	0	0	44
10%	1/1	200	1090	820	915	753	0	16
		300	1040	758	465	0	0	55
		400	1090	850	550	0	0	49
20%	1/1	205	940	1060	800	942	0	23
		305	940	1060	560	0	190	46
		390	940	1060	645	0	420	38
		460	940	1060	880	0	330	15
20%	1/1	200	1070	780	770	294	0	28
		300	1020	780	580	0	180	43
		385	1025	780	690	0	220	33
		460	1025	780	960	0	165	6
5%	3/1	200	990	1095	850	1059	0	14
		300	990	1095	730	848	0	26
		395	990	1095	365	340	0	63
		455	990	1095	305	0	0	69
5%	3/1	220	1050	780	920	706	0	12
		300	1050	780	760	118	0	28
		400	1050	780	465	0	0	56
		455	1050	780	425	0	0	57

(continued)

TABLE 4 (cont.)

Catalyst Active Material	Fe-Cr	Temp. (°C)	Inlet Gas		Outlet Gas			No Conversion (%)
			NO (ppm)	NH_3 (ppm)	NO (ppm)	NH_3 (ppm)	N_2O (ppm)	
10%	3/1	200	1055	1106	820	824	0	22
		300	1120	1106	465	259	0	58
		390	1120	1106	395	0	150	65
		450	1120	1106	560	0	0	48
10%	3/1	200	1100	735	815	530	0	26
		300	1100	735	520	118	0	52
		400	1100	735	530	0	0	52
		445	1100	735	640	0	0	42
20%	3/1	210	1095	1140	600	918	0	40
		300	910	1140	335	153	0	63
		390	910	1140	690	0	400	24
		450	910	1140	730	0	N.A.	20
		465	1110	1140	900	0	140	19
20%	3/1	200	1000	780	780	563	0	22
		300	1000	780	480	0	140	52
		400	1000	780	665	0	200	34
		465	1000	780	975	0	190	3
5%	9/1	200	1110	1120	965	1059	0	13
		300	1110	1120	805	753	0	27
		395	1110	1120	335	0	100	70
		475	1110	1120	425	0	100	62
5%	9/1	200	1130	763	965	659	0	15
		310	1130	763	690	553	0	39
		400	1130	763	430	0	0	62
		470	1130	763	510	0	0	55
10%	9/1	215	975	1106	865	965	0	11
		300	980	1106	410	448	0	58
		400	970	1152	235	0	150	76
		460	970	1152	450	0	0	54
10%	9/1	210	1000	750	825	636	0	18
		300	1000	750	480	165	0	52
		400	1000	750	515	0	0	49
		460	1000	750	605	0	0	40

(continued)

TABLE 4 (cont.)

Catalyst Active Material	Fe-Cr	Temp. (°C)	Inlet Gas NO (ppm)	Inlet Gas NH_3 (ppm)	Outlet Gas NO (ppm)	Outlet Gas NH_3 (ppm)	Outlet Gas N_2O (ppm)	NO Conversion (%)
20%	9/1	200	1010	1118	725	636	0	28
		295	1010	1118	225	330	0	78
		390	1010	1118	475	118	200	53
		300	1010	1118	215	NA	0	79
		455	1010	1118	765	0	200	24
20%	9/1	210	940	742	680	378	0	28
		300	940	753	395	0	0	62
		400	940	753	570	0	90	45
		455	940	753	745	0	130	28
		300	940	742	335	0	0	64

s.v. = 20,000 hr^{-1}. Simulated flue gas contains also 12% CO_2, 3% O_2, 5% H_2O in N_2.

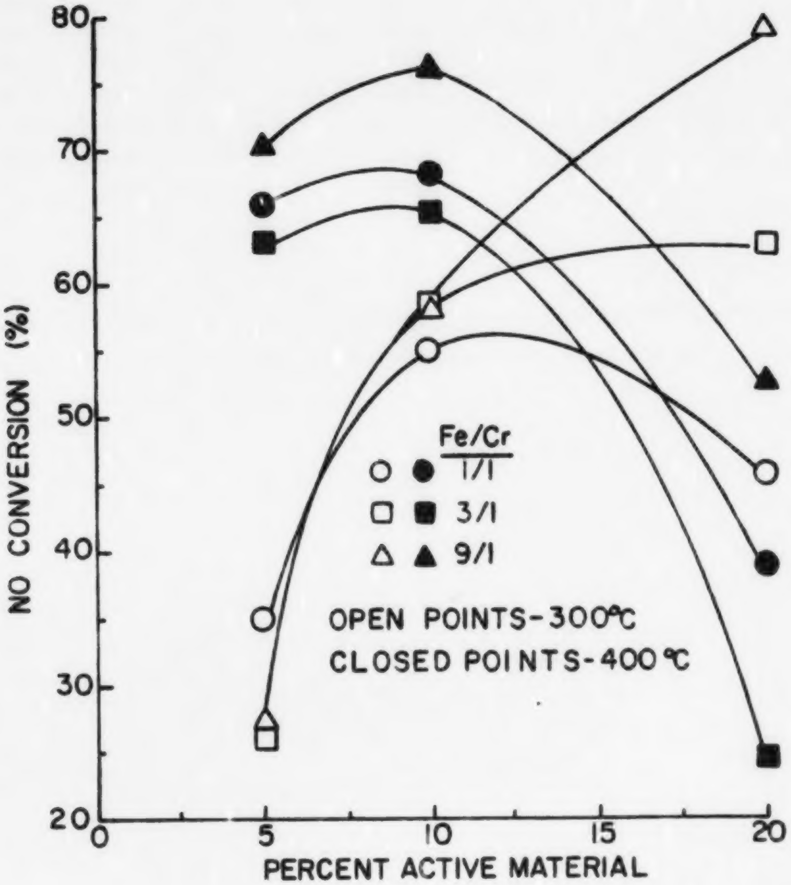

Figure 12. Effect of Fe-Cr ratio and percent active material on NO conversion at 300°C and 400°C. 1000 ppm NO, 1100 ppm NH_3. (s.v.=20,000hr^{-1}).

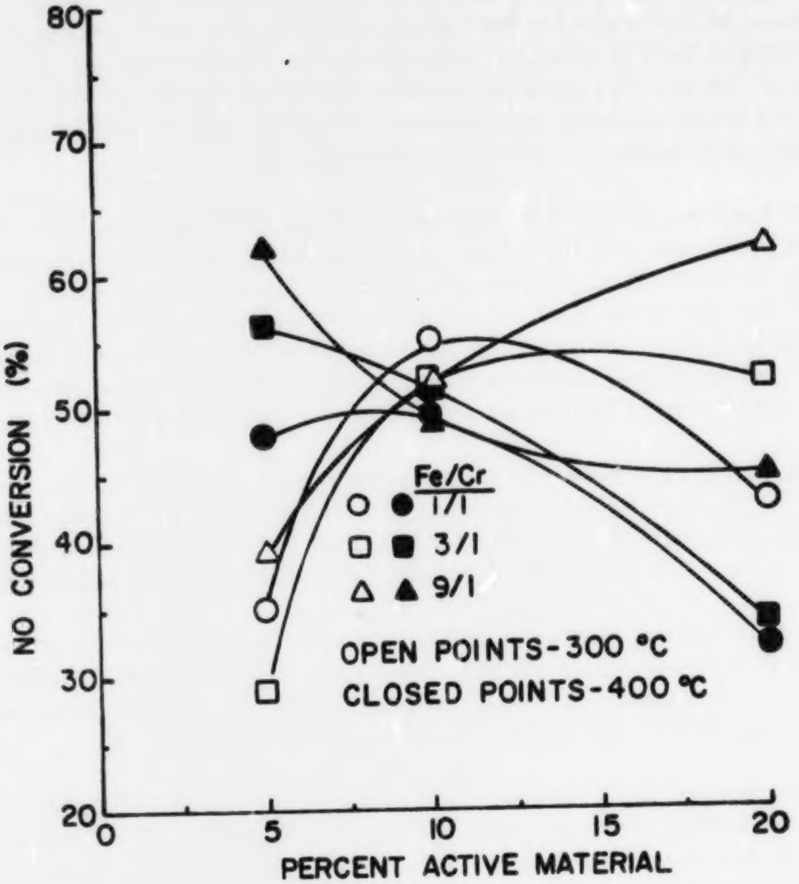

Figure 13. Effect of Fe-Cr ratio and percent active material on NO conversion at 300°C and 400°C. 1000 ppm NO, 700-850 ppm NH_3. (s.v.=20,000 hr^{-1}).

complete economic balance between NH_3 usage and required NO_x abatement (at a particular site) may indicate acceptable operation at near-stoichiometric NH_3 levels. However, to select the optimum catalyst for the parametric tests, the primary criterion employed in this phase was maximum conversion of NO. Thus, the data in Figure 12 was used to select the catalyst for operation with excess NH_3.

Figure 12 shows that with Fe/Cr ratio of 9, conversion of NO for the 20% active material at 300°C and for the 10% active material at 400°C is approximately equivalent. Selection of the 20% catalyst would depend on whether cost savings occur by incorporation of flue gas diversion ductwork at 300°C rather than at 400°C (i.e., less insulation and lower operating temperature for the fly-ash removal cyclone), and if such savings compensate for the higher catalyst cost.

It has been shown (1-2) that operation at 400°C is amenable to existing economizer outlet conditions with little heat loss. Operation at 300°C will probably require significant alteration in air preheater conditions (normal flue gas outlet temperatures of 180°C are common), such as initial cooling by air to 300°C and then final heat recovery by further cooling after catalytic conversion to desired stack gas temperatures. Thus, potential cost saving inherent in lower-temperature ductwork (less insulation, etc.) may be more than offset by additional equipment modifications.

It should also be noted (Table 4) that the 20% oxide catalysts promote the formation of N_2O; its effect on air quality has not yet been fully assessed. Similarly, the data indicate that the 20% catalysts do not appear to be as efficient in decomposing (or oxidizing) excess NH_3 at 300°C compared to the 10% catalysts at 400°C.

These several factors suggested the tentative catalyst selection of the 10% active material and operation at 400°C. However, Figure 12 suggests that even higher improvement in activity may be possible if the Fe/Cr ratio is increased to values higher than the maximum (9/1) employed in the preliminary survey.

In a second series of tests, catalysts containing Fe/Cr ratios of 20 and 50, both at a 10% loading, were prepared and tested. Figure 14 summarizes the results. Table 5 identifies the catalysts represented. In addition, catalysts containing Cr-free iron oxide were studied - 10% Fe_2O_3 on Al_2O_3, 20% Fe_2O_3 on Al_2O_3, and commercial 10% Fe_2O_3 on Al_2O_3 (Harshaw Fe-0301-T 1/8). Finally, to complete the test matrix, the Fe-Cr composition examined in the previous program (with TRW) was retested. This catalyst was about 11% active material on Al_2O_3 and contained an Fe/Cr ratio of 5.

Figure 14 shows that the commercial Fe_2O_3 catalyst has significantly lower activity than the laboratory-prepared catalysts. The former catalyst and the two prepared Fe_2O_3 catalysts (10% and 20% Fe_2O_3) appear to exhibit maximum conversion at temperatures somewhat higher than 400°C, which was the typical maximum temperature of the Fe-Cr series. Included in the figure are data points for the 5/1 Fe-Cr

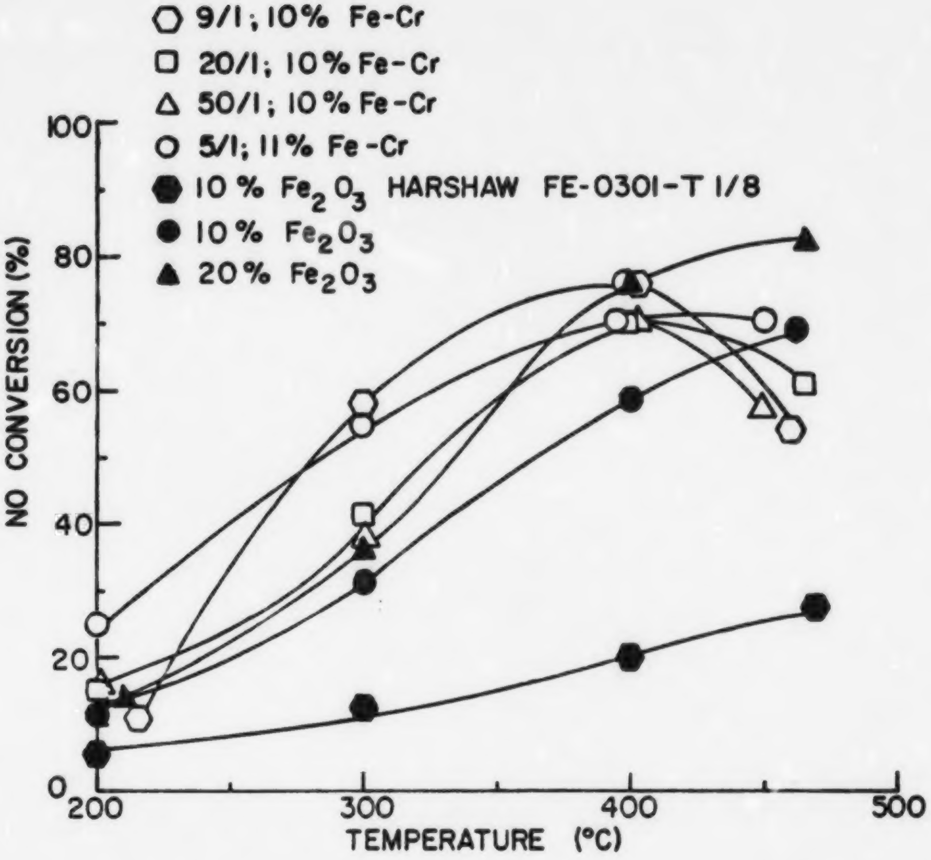

Figure 14. Conversion of NO with Fe and Fe-Cr catalysts in simulated flue gas. (s.v.=20,000 hr^{-1}). 1000 ppm NO, 1100 ppm NH_3.

TABLE 5. CONVERSION OF NO AT 400°C ON Fe AND Fe–Cr CATALYSTS

Catalyst Number	Active Material (%)	Fe/Cr Ratio (wt)	% Fe_2O_3	% Cr_2O_3	Conversion of NO at 400°C (%)
1	5	1	2.5	2.5	66
2	5	3	3.7	1.3	63
3	5	9	4.5	0.5	70
4	10	1	4.9	5.1	68
5	10	3	7.5	2.5	66
6	10	9	9.0	1.0	76
7	10	20	9.5	0.5	70
8	10	50	9.8	0.2	70
9	10	∞	10.8	0	59
10	11	5	10.6	0.4	70
11	20	1	9.9	10.1	36
12	20	3	14.9	5.1	24
13	20	9	18.0	2.0	53
14	20	∞	20.0	0	76

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s.v. = 20,000 hr^{-1}. 1000 ppm NO, 1100 ppm NH_3, 12% CO_2, 3% O_2, 5% H_2O in N_2.

catalyst (No. 10) obtained in the previous program and the 9/1 Fe-Cr catalyst (No. 6), which show that at 400°C with a 10% catalyst there is a definite increase in activity when a small amount of chromium is added to Fe_2O_3. The 20/1Fe/Cr, 50/1 Fe/Cr and 5/1Fe/Cr catalysts are of equivalent activity. The 20% Fe_2O_3 catalyst is distinctly superior to 10% Fe_2O_3 and, at 400°C, is equivalent to the Fe-Cr catalysts with 10% loading. It is apparent from Figure 14 that optimum operation of the pure Fe_2O_3 catalysts will require temperatures of 450° or higher.

It is considered that 400°C is the maximum desirable temperature for operation of the NO_x catalysts. Table 5 and Figure 15 summarize NO conversion at that temperature as a function of absolute Fe_2O_3 and Cr_2O_3 concentrations in order to assist in the selection of the optimum composition. The marked improvement in activity near 10% loading by addition of small amounts of Cr_2O_3 is shown in the plot in the direction perpendicular to the X_{NO}-Fe_2O_3 plane from point 9 to point 6.

An initial, cursory examination of the results may suggest that the single-component, 20% Fe_2O_3 formulation is the most economical catalyst. However, a more careful examination indicates that a catalyst of equivalent activity can be prepared by starting with the 20% Fe_2O_3 formulation and substituting over half of the Fe_2O_3 content with an amount of Cr_2O_3 equal to only 9% of the Fe_2O_3 replaced (i.e., the catalyst weight of active material is decreased from 20% Fe_2O_3 to a 10% catalyst with Fe/Cr = 9/1). By assuming that commercial quantities of catalyst are prepared following the procedure currently employed in the laboratory and assigning an arbitrary cost of 100 to the 20% Fe_2O_3 catalyst, the calculated relative cost of the 9/1 Fe-Cr catalyst with a 10% loading would be 45. The relative cost is based on current, moderate-volume prices for $Fe(NO_3)_3 \cdot 9\,H_2O$ and CrO_3.

The only support material other than Filtrol Grade 86 that was investigated in this project with the Fe-Cr catalysts was the Aeroban Al_2O_3-SiO_2 which was shown to be similar to the Filtrol Al_2O_3 with V_2O_5 loading. Tests were performed to determine if the Aeroban substrate is an equivalent or superior carrier (compared with Filtrol Grade 86) for the 9/1 Fe-Cr catalyst selected for parametric testing.

Figure 16 shows that in simulated flue gas (12% CO_2, 5% H_2O and 3% O_2 in N_2), conversion of 1000 ppm NO with 1100 ppm NH_3 is significantly higher on the catalyst with the Filtrol support. Conversion of NO maximized at 300°C with Aeroban, compared to about 400°C for Filtrol support. The low net conversion of NO on Aeroban appears to be related to enhanced preferential removal of NH_3 by oxidation with O_2 or by decomposition; the NH_3 conversion rate was substantially higher on the Aeroban catalyst as shown in the figure. The Aeroban-supported catalyst apparently promotes the formation of NO by oxidation of NH_3 with O_2 at substantially lower temperatures than the Filtrol catalyst.

The only commercial Fe-Cr catalyst tested during this project was Girdler G3A. Preliminary tests with this catalyst (87% oxides with Fe/Cr ratio of 12 on MgO +

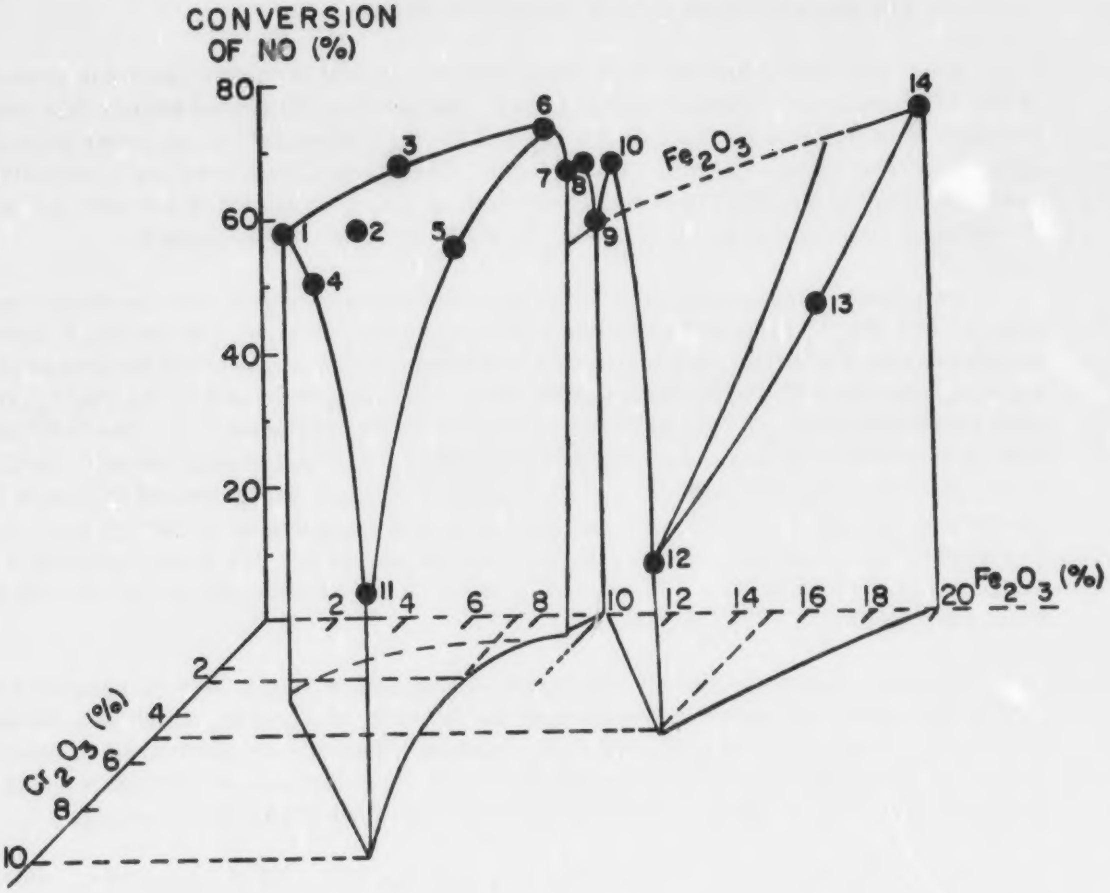

Figure 15. Conversion of NO as a function of Fe_2O_3 and Cr_2O_3 concentrations at 400°C. See Table 5 for catalysts denoted by numbered points. (s.v.= 20,000 hr^{-1}).

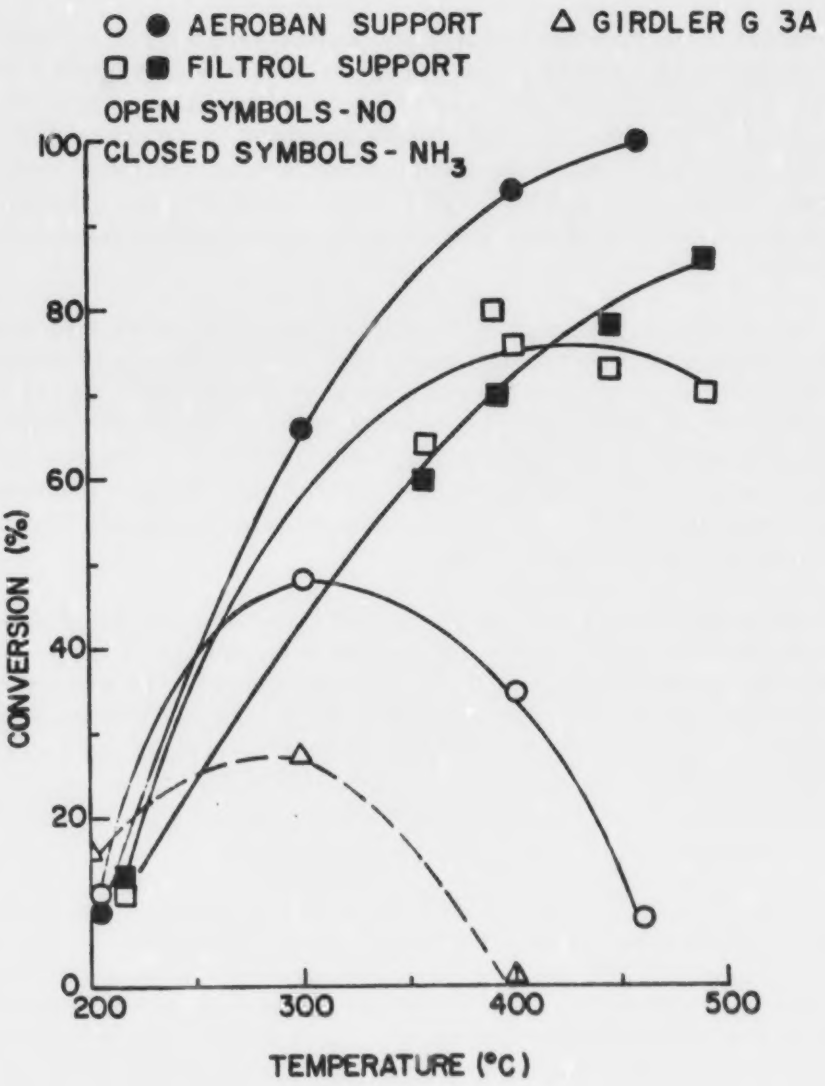

Figure 16. Conversion of NO and NH_3 on 10% Fe-Cr catalysts in simulated flue gas. 1000 ppm NO, 1100 ppm NH_3. (s.v.=20,000 hr^{-1}).

SiO_2 + graphite) in the previous UCLA/TRW project were not particularly promising, based on a single point screening test. Nevertheless, it was of interest to determine the activity of this Fe-Cr catalyst under present test conditions. In particular, it was considered possible that the temperature employed in the previous test (400°C) may not have been that at which the maximum conversion of NO was realized with this catalyst. Previously, at 400°C with stoichiometric NH_3 in simulated flue gas, conversion of NO was only 2% and very extensive decomposition (or oxidation) of NH_3 was observed.

Tests of the Girdler catalyst with stoichiometric NH_3 were conducted in the current project at temperatures varying from 200 to 400°C. Figure 16 includes these results. At 400°C conversion of NO was zero even though total usage of NH_3 was observed. At 300°C, at which point the NO conversion maximum was observed, NO conversion was only 26%. Considerable N_2O was produced, in contrast to the Fe-Cr-Al_2O_3 catalysts prepared at UCLA, which appeared to be formed primarily from the reaction between NH_3 and O_2. No further tests were made with the G3A catalyst completing the catalyst optimization studies.

Surface areas of several of the Fe-Cr catalysts were measured to determine if activity was related to area. Table 6 summarizes the results of the measurements and includes NO conversion data at 400°C. There appears to be a correlation between area and activity for the Fe-Cr catalysts although the deviation in areas of the three catalysts are not markedly different (values given are averages of several measurements, generally within ± 5 sq m/gm). Conversion of NO on the two chromium-free catalysts appeared to be more dependent on the amount of Fe_2O_3 than on surface area as shown in the table.

Based on the activity tests, the 10% catalyst with an Fe/Cr ratio of 9/1 on Filtrol Grade 86 support was selected for further parametric tests. At that time there was no reason to expect that the SO_x resistance would be significantly different from the 11% , Fe-Cr (5/1) catalyst, which had been previously shown to be unaffected by SO_x at 400°C for short exposure time tests. Nevertheless, parametric tests of the 9/1 Fe-Cr catalyst were preceded by a short experimental study of the effect of SO_x on conversion of NO on the 9/1 Fe-Cr catalyst in simulated flue gas. For short-term exposure to SO_x (8 hours) there was no evident deterioration in catalyst activity; (see Table 7) thus, a final selection of the 9/1 Fe-Cr catalyst for use in further parametric studies was made.

5.1.3. Preparation of Optimum Catalyst Formulations

The laboratory procedures used in the preparation of the 15% V_2O_5 and the 10% Fe-Cr (9/1) catalyst are described in this section. The selected carrier was Filtrol Grade 86 Al_2O_3 pellets (~1/8 in. dia. x 3/16 in. long).

The V_2O_5 catalyst was prepared by impregnating 85 gm of dry Al_2O_3 pellets

TABLE 6. SURFACE AREA OF Fe-Cr CATALYSTS

Fe/Cr Ratio (wt. basis)	Oxide Content (%)	Area (sq m/gm)	Conversion of NO at 400°C (%)
3	10	240	66
9	10	260	76
50	10	240	70
∞	10	240	59
∞	20	200	76
Raw Filtrol Carrier	0	270	--

TABLE 7. EFFECT OF SO_2 ON CONVERSION OF NO OVER Fe-Cr (9/1) CATALYST

Inlet Gas Conc.			Outlet Gas Conc.			Conv. of
NO (ppm)	NH_3 (ppm)	SO_2 (ppm)	NO (ppm)	NH_3 (ppm)	Temp. (°C)	NO (%)
900	1317	0	675	790	295	25
900	1323	800	690	565	295	24
890	1317	0	240	400	400	73
890	1323	800	250	56	400	72

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s.v. = 20,000 hr^{-1}. Simulated flue gas contains also 12% CO_2, 3% O_2, 5% H_2O in N_2.

with 60 ml of a solution containing 19.3 gm NH_4VO_3 and 10.4 gm $H_2C_2O_4 \cdot 2H_2O$ (2:1 molar ratio). Complete absorption of the solution could not be achieved in one step. The excess liquor was decanted from the pellets. The latter were then dried in ambient air and subsequently impregnated with the decanted liquor.

The Fe-Cr catalyst was prepared by impregnating 90 gm of the dry alumina pellets with a 64 ml solution of 45.5 gm $Fe(NO_3)_3 \cdot 9H_2O$ and 1.3 CrO_3.

Both the V_2O_5 - Al_2O_3 and Fe-Cr-Al_2O_3 catalysts were calcined in flowing air at 500° C for 16 hours. Calcination burned off the organic acid and decomposed the vanadate to V_2O_5, decomposed the ferric nitrate and oxidized Cr(III) to Cr(VI).

5.1.4. Summary of Catalyst Physical Properties

Table 8 summarizes the physical characteristics of the catalysts selected for further study in the parametric and SO_2 durability studies. The pertinent parameters are surface area, pore volume, bulk and particle density and average pore radius. Pore volumes were measured by acetone absorption and bulk densities by mercury displacement. BET surface areas were determined by N_2 adsorption at the normal b.p. of liquid nitrogen. Average pore radii were calculated from measured surface areas and pore volumes.

TABLE 8. CATALYST PHYSICAL PROPERTIES

Catalyst	Surface Area (sq. m/gm)	Pore Volume (cu cm/gm)	Porosity (%)	Bulk Density (gm/cu cm)	Particle Density (gm/cu cm)	Av. Pore Radius (A)
Filtrol Grade 86 Support	270	0.68	70	1.02	3.38	51
15% V_2O_5	220	0.48	58	1.21	2.90	44
10% Fe-Cr (9/1)	260	0.60	69	1.16	3.72	45

5.2. Task 2. PARAMETRIC INVESTIGATION OF VANADIA AND IRON-CHROMIUM CATALYSTS IN THE PRESENCE OF SO_x

In addition to the determination of the effect of the presence of SO_x on the temperature-activity characteristics of V_2O_5 and Fe-Cr catalysts during short and long-term exposure, as stated in the title of this study task, a concurrent objective was to develop rate expressions for the catalysts suitable for design use. The rate expressions should represent intrinsic surface rates, including the effects of O_2, H_2O and CO_2 as well as SO_x, so that reasonable predictions of performance over a wide range of operating conditions can be made.

The approach used in this task was to vary widely the operating parameters about the standard flue gas conditions (12% CO_2, 3% O_2, 5% H_2O, 1000 ppm NO, stoichiometric or excess NH_3 in N_2 at 400° C and 20,000 hr^{-1} (STP) space velocity. The parameters were varied individually or in various combinations. After establishing tentative performance characteristics in the absence of SO_x, the test procedure was repeated, in part, with SO_x added to the flue gas. The data were correlated in the development of empirical rate expressions.

At the conclusion of the parametric test program each of the catalysts was subjected to long-term exposure to SO_x under operating conditions. The important parameters, such as temperature, NH_3/NO ratio, and space velocity were varied during the durability test program.

5.2.1. Parametric Studies

The results of previous studies and those obtained in Task 1 indicated that conversion of NO in flue gas was the result of several parallel and consecutive processes. Both V_2O_5 and Fe-Cr catalysts exhibited maxima in conversion as a function of temperature and both were sharply influenced by the presence of O_2.

For design use, it is desirable that working rate expressions be as simple as possible. To meet this objective, power law rate expressions were used to correlate the experimental kinetic data. The effects of CO_2, H_2O, O_2 could be incorporated as individual concentration-reaction order terms. A thorough kinetic evaluation involving surface phenomena and reaction mechanisms to determine the role of each species was deemed beyond the scope of this project.

The NO-NH_3 reaction is sufficiently complex that the determination of a single, simple rate expression which will be valid over entire spectrum of possible flue gas conditions is not likely. The rate of NO reduction on both catalysts was markedly increased by increases in O_2 content at low O_2 levels. Above about 0.5% the conversion of NO was not as markedly increased with increase in O_2. Depending on temperature, O_2 may have either an enhancement or an inhibition effect on removal of NO. Although the influence of H_2O and CO_2 on the rate of reaction was not known, the exhaust of fossil fuel-burning plants will never be devoid of CO_2 or H_2O. Therefore, it is reasonable to examine only the expected concentrations of these species for fuels ranging from coal to natural gas. In the earlier program, the presence of SO_x at concentrations of 1000-1500 ppm did not appear to influence the reduction of NO in excess NH_3. However, a poisoning term may be required in the rate expressions at higher SO_x concentrations.

The most difficult task is the mathematical representation of the rate of reduction of NO near and above the temperature of maximum conversion. Two approaches were considered. First, simple separate rate expressions for apparent conversion of NO may be developed to define the ascending and descending portions of the conversion curve. Second, since the maximum in the conversion of NO is probably caused by the onset of NH_3 oxidation with subsequent depletion of the reducing agent (and, frequently, the actual production of NO), it may be possible to combine a single expression for the rate of reduction of NO with a parallel rate expression for the oxidation of NH_3.

The experimental approach that was followed in the parametric tests was to first use the standard simulated flue gas mixture typical for coal combustion (12% CO_2, 5% H_2O, 3% O_2) varying the NO concentration from 250 to 1500 ppm and the NH_3 concentration between about 0.5 to 1.5 times the stoichiometric requirement for each NO concentration. Then, at the maximum conversion point with each NO/NH_3 ratio, the CO_2 and H_2O concentrations were perturbed from the standard flue gas concentrations to the maximum and minimum values expected in actual use (e.g., from about 8% to 15% CO_2 and from about 3% to 18% H_2O). Also, additional tests were conducted with gas containing neither CO_2 nor H_2O (for use as a reference base).

The effect of changes in the O_2 concentration were investigated at concentrations of NO of 750 ppm and 1000 ppm with at least two concentration levels of NH_3 (stoichiometric and 1.5 times stoichiometric) and oxygen concentrations varying from 0 to 5 %. In these tests, temperatures were varied from 280 to 500°C.

The last phase of the parametric tests included the addition of SO_x at various concentrations to the test gas. In this latter series of tests, the possible cross effects with H_2O, O_2 and NH_3 were assessed by including SO_x in a test matrix.

5.2.1.1. V_2O_5 Catalyst

Table 9 presents typical preliminary parametric data obtained for the V_2O_5 catalyst at 20,000 hr^{-1} space velocity for variations in temperature and concentrations of NO_x, NH_3 and O_2. The following tentative conclusions are indicated from the data:

1. Nitrous oxide (N_2O) is not a significant product with the V_2O_5 catalyst except at high concentrations of NO and NH_3 and then only at temperatures well above that for maximum conversion (conversion maximized at 400°C).

2. Conversion of NH_3 continues to increase monotonically with temperature beyond the point of maximum NO conversion.

3. The NH_3 concentration influences NO conversion most strongly below about 1.5 times the stoichiometric level (i.e. NH_3/NO inlet concentration ratio of 1). Above this concentration ratio, excess NH_3 does not influence the reaction substantially.

4. Varying the H_2O concentration from 0 to 18% does not have a significant influence on the conversion of NO.

TABLE 9. PARAMETRIC STUDIES TEST DATA - V_2O_5 CATALYST

Inlet Gas Composition					Outlet Gas Comp.			Temp.	Conversion of	
NO_x	NH_3	CO_2	H_2O	O_2	NO_x	N_2O	NH_3		NO	NH_3
(ppm)	(ppm)	(%)	(%)	(%)	(ppm)	(ppm)	(ppm)	(°C)	(%)	(%)
290	186	12	5	3	225	0	0	200	22	100
295	"	"	"	"	140	0	0	300	52	100
"	194	"	"	"	110	0	0	400	59	100
285	186	"	"	"	185	0	0	500	35	100
290	248	12	5	3	255	0	140	200	28	43
280	245	"	"	"	105	0	47	300	63	81
"	"	"	"	"	75	0	0	400	73	100
"	"	"	"	"	140	0	0	490	50	100
275	350	12	5	3	200	0	187	200	27	47
"	"	"	"	"	105	0	0	300	62	100
"	"	"	"	"	60	0	0	395	78	100
"	"	"	"	"	145	0	0	490	47	100
280	140	12	5	3	270	0	105	200	4	25
"	"	"	"	"	255	0	70	300	9	50
"	"	"	"	"	245	0	0	400	13	100
"	"	"	"	"	255	0	0	490	9	100
285	315	12	5	3	175	0	163	205	39	48
"	"	"	"	"	65	0	0	305	77	100
"	"	"	"	"	40	0	0	395	86	100
295	314	"	"	"	140	0	0	495	53	100
770	445	12	5	3	585	0	301	200	24	34
"	"	"	"	"	295	0	175	300	62	62
"	"	"	"	"	280	0	82	400	64	82
"	"	"	"	"	375	0	0	500	51	100
805	536	12	5	3	630	0	373	200	22	50
"	501	"	"	"	315	0	140	300	61	72
"	"	"	"	"	315	0	47	400	61	91
"	"	"	"	"	440	0	0	495	45	100
785	769	12	5	3	580	0	536	200	26	30
"	"	"	"	"	235	0	303	305	70	61
"	"	"	"	"	155	0	70	400	80	91
"	"	"	"	"	280	0	0	500	64	100
770	921	12	5	3	565	0	594	200	27	36

(continued)

TABLE 9 (cont.)

Inlet Gas Composition					Outlet Gas Comp.			Temp.	Conversion of	
NO_x	NH_3	CO_2	H_2O	O_2	NO_x	N_2O	NH_3		NO	NH_3
(ppm)	(ppm)	(%)	(%)	(%)	(ppm)	(ppm)	(ppm)	(°C)	(%)	(%)
770	1189	12	5	3	560	0	851	200	27	28
"	"	"	"	"	215	0	676	300	72	43
"	"	"	"	"	100	0	466	395	87	61
"	"	"	"	"	205	0	140	495	73	88
975	524	12	5	3	715	0	396	200	27	24
"	"	"	"	"	405	0	140	300	58	73
"	"	"	"	"	370	0	0	400	62	100
"	"	"	"	"	535	0	0	490	45	100
980	583	12	5	3	705	0	350	200	28	40
"	"	"	"	"	425	0	128	300	57	78
"	"	"	"	"	405	0	70	400	59	88
"	"	"	"	"	537	0	0	495	45	100
980	957	12	5	3	710	0	641	200	28	33
"	967	"	"	"	340	0	326	300	65	66
"	"	"	"	"	205	0	198	400	79	80
"	"	"	"	"	385	0	70	495	61	93
985	1259	12	5	3	720	0	758	200	27	40
"	"	"	"	"	275	0	513	300	72	59
"	"	"	"	"	160	0	303	400	84	76
"	"	"	"	"	305	0	140	495	69	89
1005	1480	12	5	3	735	0	1270	200	27	14
995	1468	"	"	"	275	0	816	300	72	44
"	"	"	"	"	105	0	629	395	89	57
"	"	"	"	"	275	0	221	495	72	85
1460	781	12	5	3	1100	0	455	200	24	42
"	"	"	"	"	740	0	150	300	49	81
"	"	"	"	"	675	0	0	400	54	100
"	"	"	"	"	815	0	0	500	44	100
1475	1002	12	5	3	1090	0	466	200	26	53
1460	967	"	"	"	645	0	210	300	56	78
"	"	"	"	"	545	0	93	395	63	90
"	"	"	"	"	785	0	0	495	46	100
1410	1457	12	5	3	1040	0	1084	200	26	26
"	"	"	"	"	460	0	548	300	67	62

(continued)

TABLE 9 (Cont.)

Inlet Gas Composition					Outlet Gas Comp.			Temp.	Conversion of	
NO_x (ppm)	NH_3 (ppm)	CO_2 (%)	H_2O (%)	O_2 (%)	NO_x (ppm)	N_2O (ppm)	NH_3 (ppm)	(°C)	NO (%)	NH_3 (%)
1485	1980	12	5	3	1090	0	1755	200	27	11
1475	1980	"	"	"	435	0	1165	300	71	41
"	"	"	"	"	225	0	635	395	85	68
"	"	"	"	"	485	100	233	495	67	88
1485	2400	12	5	3	1070	0	1865	200	28	22
"	"	"	"	"	415	0	1492	300	72	38
"	"	"	"	"	175	0	1165	400	88	52
"	"	"	"	"	370	100	350	500	75	85
1025	676	12	5	3	395	0	0	400	62	100
"	"	8	"	"	370	0	0	"	65	"
"	"	14	"	"	390	0	0	"	62	"
1070	"	12	10	"	405	0	0	"	62	"
985	"	"	0	"	335	0	0	"	66	"
1030	1480	12	5	3	95	0	338	400	91	77
"	"	8	"	"	105	0	326	"	90	78
990	1503	14	"	"	140	0	536	"	86	64
1000	"	12	10	"	135	0	338	"	87	77
960	"	"	0	"	85	0	299	"	91	80
850	1165	12	5	3	155	0	443	400	82	62
"	"	8	"	"	140	0	456	"	84	61
"	"	14	"	"	165	0	443	"	81	62
965	1165	12	10	"	"	0	"	"	83	"
970	"	"	0	"	105	0	"	"	89	"
950	734	12	5	3	245	0	93	400	74	87
"	"	8	"	"	225	0	186	"	76	75
"	"	14	"	"	235	0	105	"	75	86
1000	"	12	10	"	"	0	163	"	77	78
985	"	"	0	"	165	0	93	"	83	87
955	"	"	0	"	175	0	116	"	82	84
990	1442	12	5	5	105	0	559	400	89	61
"	"	"	"	3	135	0	513	"	86	64
"	"	"	"	1	167	0	606	"	83	58
"	"	"	"	0.5	195	0	653	"	80	55
"	"	"	"	0	845	0	1235	"	15	14

(continued)

TABLE 9 (cont).

Inlet Gas Composition					Outlet Gas Comp.			Temp.	Conversion of	
NO_x (ppm)	NH_3 (ppm)	CO_2 (%)	H_2O (%)	O_2 (%)	NO_x (ppm)	N_2O (ppm)	NH_3 (ppm)	(°C)	NO (%)	NH_3 (%)
985	1491	12	5	5	235	0	858	300	76	43
"	"	"	"	3	257	0	904	"	74	39
1050	1445	"	"	1	305	0	816	"	71	43
"	"	"	"	0.5	375	0	"	"	62.	36
"	"	"	"	0	885	0	1457	"	10	2.
990	1492	"	"	3	300	0	210	490	70	86
990	1459	"	"	3	320	0	210	500	68	86
970	1459	"	"	5	290	0	186	"	70	87
1030	1459	"	"	1	340	0	0	"	67	100
1070	1459	"	"	0.5	435	0	185	"	59	87
1070	1459	"	"	0	845	0	1014	"	21	30
990	652	"	"	3	450	0	221	300	55	66
980	652	"	"	5	405	0	210	"	59	68
1020	641	"	"	1	475	0	220	"	53	66
1025	641	"	"	0.5	525	0	198	"	51	69
1080	641	"	"	0	905	0	454	"	16	29
985	688	"	"	3	305	0	0	400	69	100
970	688	"	"	5	280	0	0	"	71	100
1020	688	"	"	1	350	0	175	"	66	75
1060	676	"	"	0.5	395	0	116	"	63	83
1075	676	"	"	0	855	0	478	"	20	29
955	699	"	"	3	475	0	0	500	50	100
970	699	"	"	5	500	0	0	"	49	100
990	699	"	"	1	535	0	47	"	46	93
1035	676	"	"	0.5	565	0	186	"	45	72
995	676	"	"	0	755	0	396	"	24	41
490	384	12	5	5	180	0	175	300	63	54
500	"	"	"	3	185	0	198	"	63	48
570	"	"	"	1	240	0	210	"	58	45
599	"	"	"	0.5	270	0	245	"	55	36
590	"	"	"	0	430	0	303	"	27	21
510	734	12	5	5	140	0	373	300	73	49
510	"	"	"	3	150	0	396	"	71	46
560	"	"	"	1	180	0	420	"	68	43
565	"	"	"	0.5	200	0	443	"	65	40
560	"	"	"	0	440	0	664	"	21	10

(continued)

TABLE 9 (cont).

Inlet Gas Composition					Outlet Gas Comp.			Temp.	Conversion of	
NO_x (ppm)	NH_3 (ppm)	CO_2 (%)	H_2O (%)	O_2 (%)	NO_x (ppm)	NH_3 (ppm)	N_2O (ppm)	(°C)	NO (%)	NH_3 (%)
490	373	12	5	5	155	0	0	400	68	100
500	"	"	"	3	170	0	0	"	66	"
520	"	"	"	1	190	105	0	"	63	72
515	"	"	"	0.5	200	"	0	"	61	"
540	"	"	"	0	320	280	0	"	41	25
495	781	12	5	5	125	326	0	400	75	58
485	"	"	"	3	130	338	0	"	73	57
525	"	"	"	1	135	361	0	"	74	54
535	"	"	"	0.5	150	385	0	"	72	51
520	"	"	"	0	275	536	0	"	47	31
490	781	12	5	5	270	93	0	500	45	88
505	"	"	"	3	265	105	0	"	48	87
530	"	"	"	1	325	140	0	"	39	82
520	769	"	"	0.5	320	158	0	"	39	79
530	"	"	"	0	330	629	0	"	38	18
480	352	12	5	5	335	0	0	500	30	100
485	"	"	"	3	370	0	0	"	24	"
520	"	"	"	1	370	0	0	"	29	"
"	"	"	"	0.5	380	47	0	"	27	87
540	"	"	"	0	365	193	0	"	32	45
810	1130	12	5	5	172	571	0	300	79	50
800	"	"	"	3	190	594	0	"	76	47
850	"	"	"	1	250	710	0	"	71	37
865	"	"	"	0.5	300	664	0	"	65	41
855	"	"	"	0	690	990	0	"	16	12
795	580	12	5	5	275	117	0	300	65	80
790	"	"	"	3	300	140	0	"	62	75
880	"	"	"	1	395	152	0	"	55	74
870	"	"	"	0.5	475	175	0	"	45	70
885	"	"	"	0	770	385	0	"	13	34
750	571	12	5	5	260	58	0	400	65	90
780	"	"	"	3	270	82	0	"	65	86
835	"	"	"	1	335	105	0	"	60	82
840	"	"	"	0.5	350	163	0	"	58	72
850	"	"	"	0	625	290	0	"	26	49

(continued)

TABLE 9 (cont.)

Inlet Gas Composition					Outlet Gas Comp.			Temp.	Conversion of	
NO_x (ppm)	NH_3 (ppm)	CO_2 (%)	H_2O (%)	O_2 (%)	NO_x (ppm)	NH_3 (ppm)	N_2O (ppm)	(°C)	NO (%)	NH_3 (%)
760	1095	12	5	5	155	408	0	400	80	63
770	"	"	"	3	160	443	0	"	79	60
800	1107	"	"	"	155	385	0	"	80	65
870	"	"	"	1	185	396	0	"	79	64
890	"	"	"	0.5	205	419	0	"	77	62
"	"	"	"	0	730	839	0	"	18	24
785	1105	12	5	5	340	117	0	495	57	89
775	"	"	"	3	367	128	0	"	53	88
860	"	"	"	1	380	152	0	"	56	86
855	"	"	"	0.5	397	163	0	"	54	85
885	"	"	"	0	635	862	0	"	28	22
750	548	12	5	3	495	0	0	495	34	100
800	"	"	"	1	515	35	0	"	36	94
815	"	"	"	0.5	545	58	0	"	33	89
"	"	"	"	0	530	198	0	"	35	64

20,000 hr^{-1} space velocity

5. The influence of CO_2 on NO conversion appears to be quite small between 8 and 14% CO_2. The largest effect observed is at stoichiometric NH_3 levels or lower and may be of only academic interest (all indications are that excess NH_3 will be required for optimum NO conversion).

6. Oxygen strongly influences the rate of NO reduction below 0.5% O_2 with either stoichiometric or excess NH_3. The effect is one of definite enhancement with excess NH_3. Interestingly, the enhancement effect is also apparent at temperatures greater than that at maximum NO conversion. Previous work in this laboratory, with platinum catalyst, indicated that O_2 inhibits the NO reaction at temperatures greater than that at maximum conversion (about 200°C with Pt).

The influence of the NH_3/NO ratio on the conversion of NO is shown clearly in Figures 17 and 18 for NO concentrations of 280 ppm and from 770 to 1485 ppm respectively. It can be seen that with an NO concentration of 770 ppm or higher (Fig. 18), the absolute NO concentration, at a given temperature, does not influence the rate of NO conversion; instead, it appears that the reactant ratio and temperature controls the conversion of NO. This observation was also made in the earlier program with Pt, V_2O_5 and Fe-Cr catalysts.

Figure 17 shows a conversion maximum at NH_3/NO ratios of about 1.1:1 at all temperatures, whereas no such behavior is evident in Figure 18. It is not known if this apparent difference in the relationship between conversion and reactant ratio for low NO concentrations indicates a change in mechanism. It is possible that uncertanties in establishing and maintaining the low flow rates may have magnified the experimental error. In fact, some difficulty was experienced in the operation of the water vaporizer, which could have led to fluctuations in the total flow (and, hence, concentrations) and back pressure.

Oxygen strongly affects NO conversion below 0.5% O_2. The O_2 effect below 0.5% O_2 is shown clearly in Figures 19, 20 and 21 for 1000 ppm, 790-890 ppm and 490-600 ppm NO, respectively. Although the nitrogen carrier gas may contain as much as 1000 ppm O_2, the data for "zero" O_2 are shown on the ordinate. It is quite likely that conversion for completely O_2-free conditions may be even lower than the values shown.

Even under "low excess air firing," O_2 concentrations are rarely below 0.5%. Furthermore, such near-stoichiometric combustion has not been fully perfected for solid fuel (coal). Therefore, the projected mathematical modelling was designed to cover O_2 concentrations above 0.5% .

Reduction of NO is enhanced by O_2 up to the temperatures of maximum conversion (400°C) for all NO levels. For 1000 ppm NO, enhancement was observed at 500°C with both NH_3 levels (Figure 19). With 790-890 ppm NO, enhancement occurs with excess NH_3 but not with stoichiometric NH_3 (Figure 20). However, with 490-600 ppm NO, no significant enhancement occurs with either NH_3 level at 500°C (Figure 21).

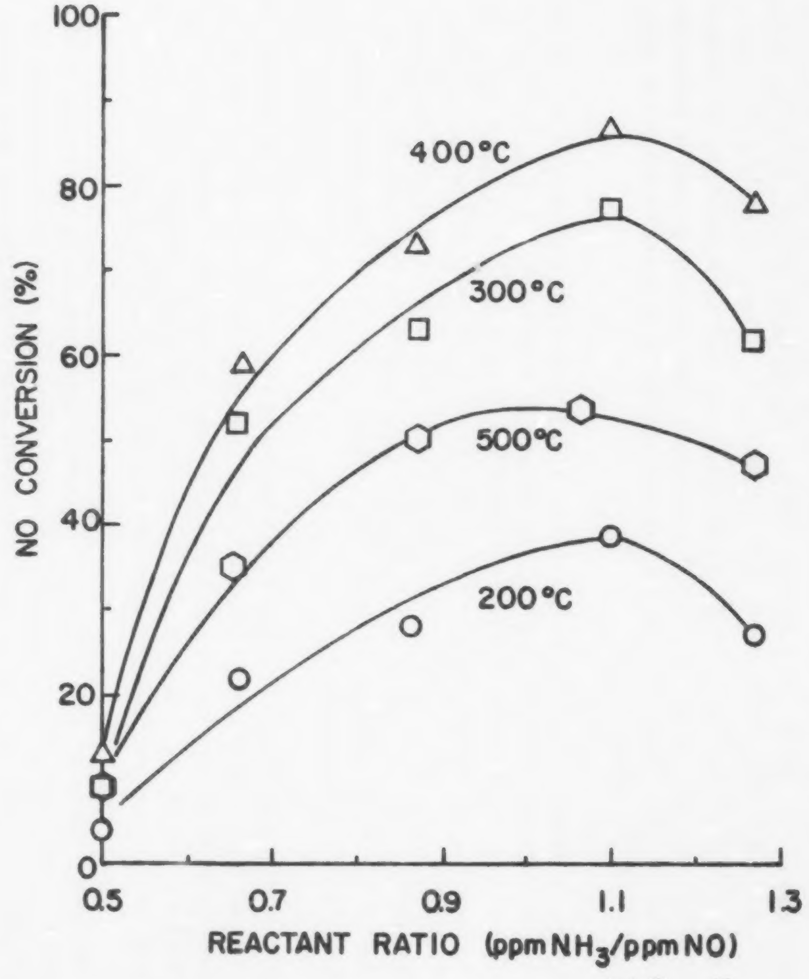

Figure 17. Removal of NO with NH_3 on V_2O_5 at low NO concentrations in simulated flue gas. 280 ppm NO. (s.v.= 20,000 hr^{-1}).

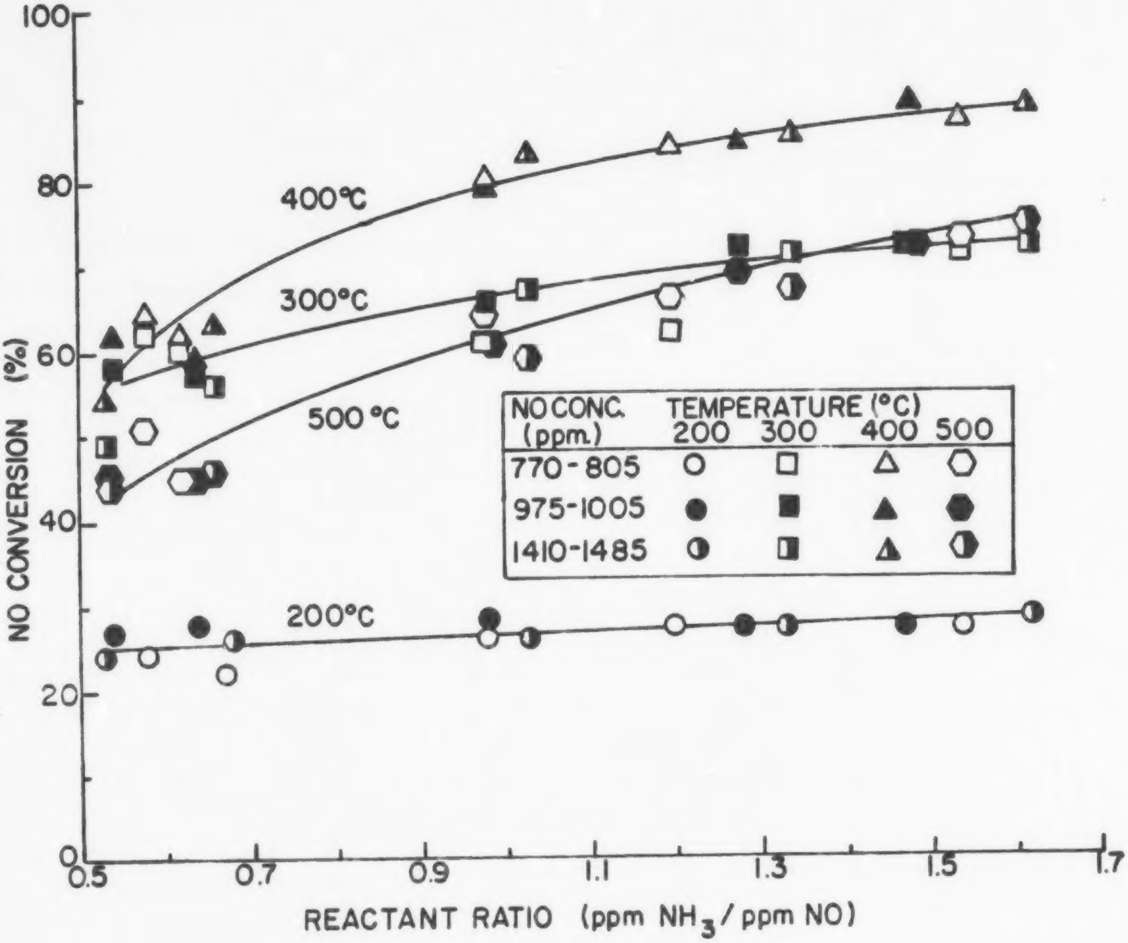

Figure 18. Removal of NO with NH_3 on V_2O_5 at high NO concentrations in simulated flue gas. (s.v.= 20,000 hr^{-1}).

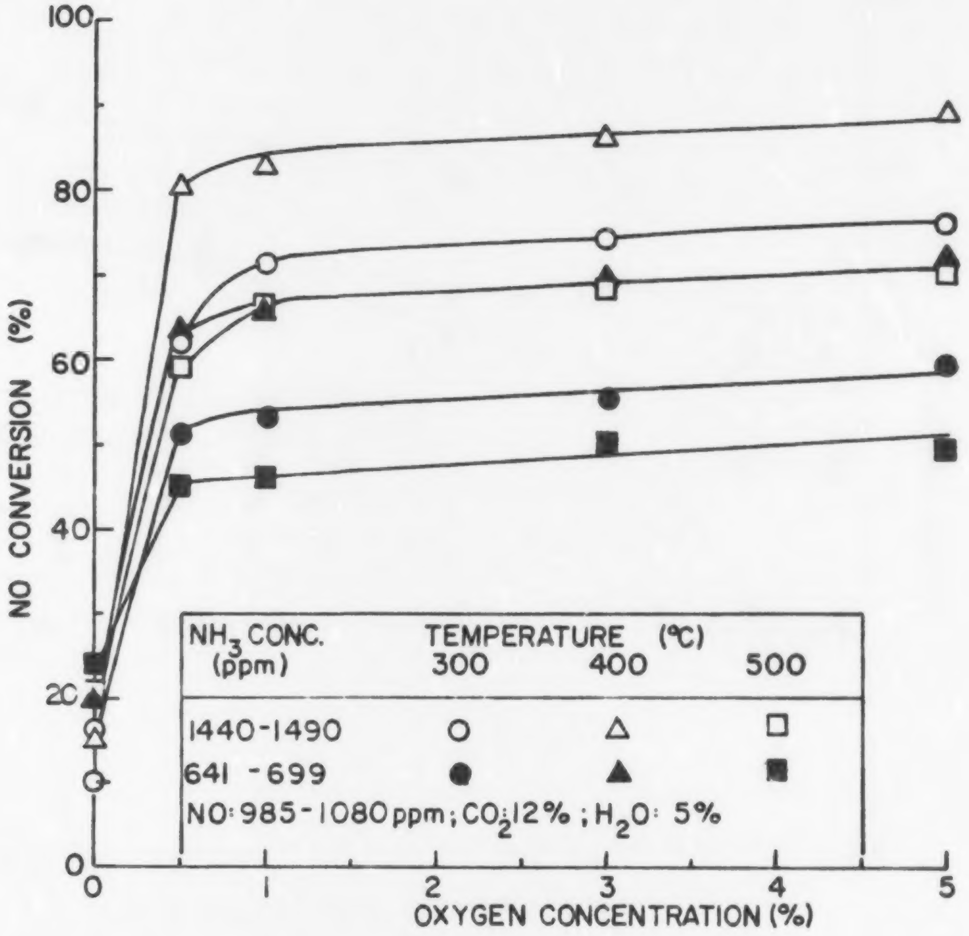

Figure 19. Effect of O_2 concentration on NO reduction on V_2O_5 in simulated flue gas at high NO concentrations. (s.v.= 20,000 hr^{-1}).

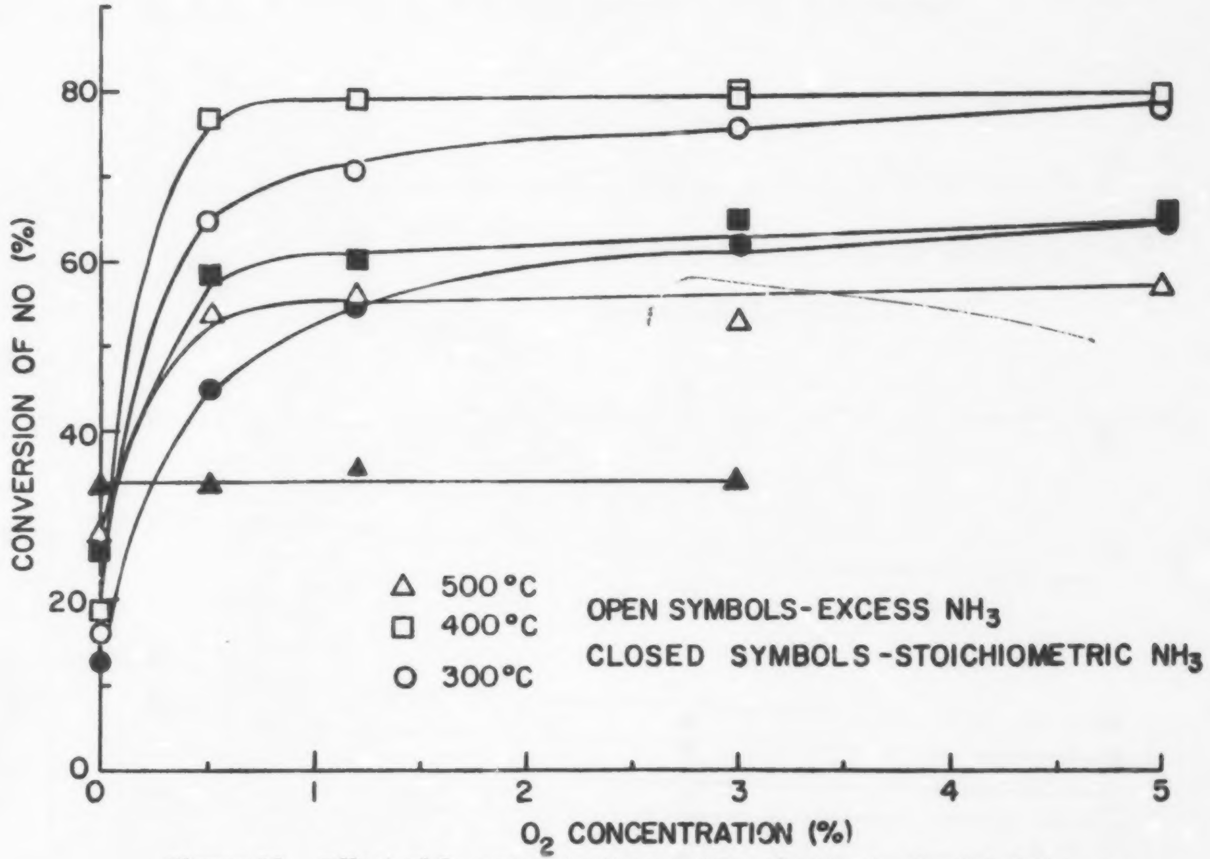

Figure 20. Effect of O_2 concentration on NO reduction in simulated flue gas on V_2O_5 catalyst at intermediate NO concentrations. 790-890 ppm NO, 12% CO_2, 5% H_2O (s.v.= 20,000 hr^{-1}).

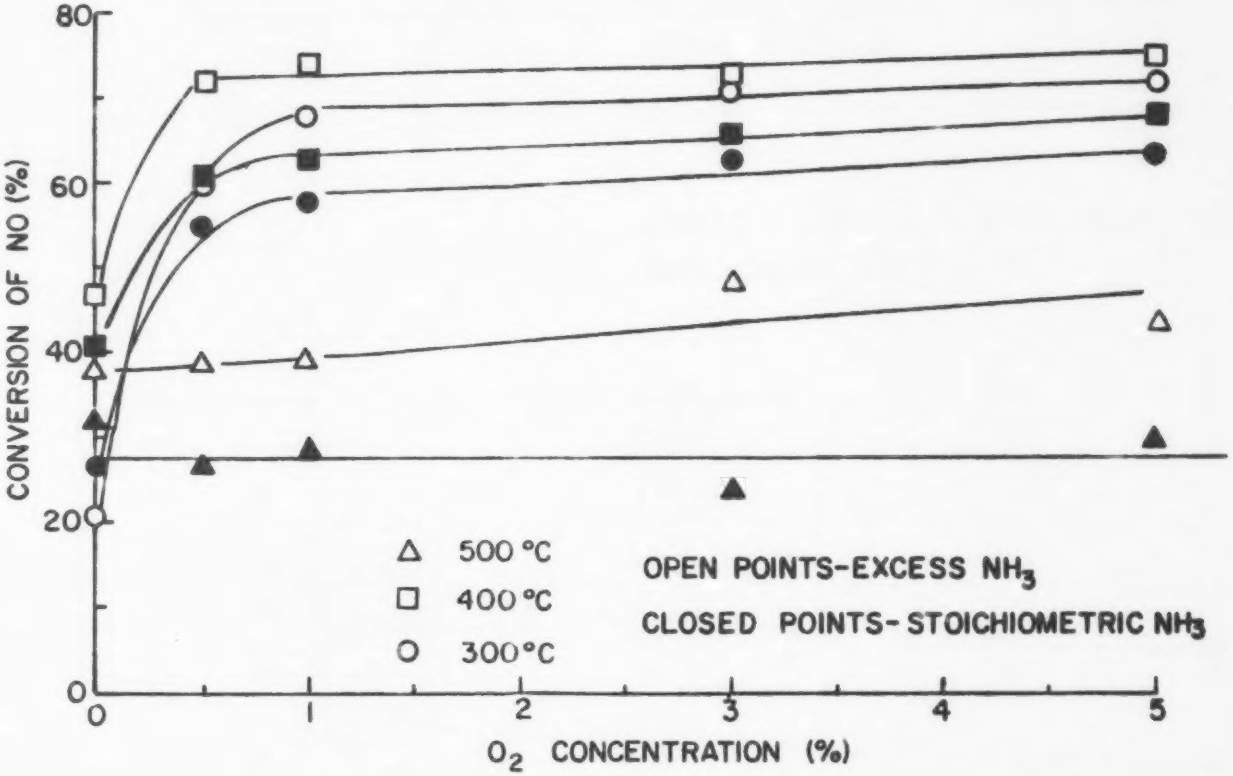

Figure 21. Effect of O_2 concentration on NO reduction on V_2O_5 in simulated flue gas at low NO concentrations. 490-600 ppm NO (s.v. = 20,000 hr^{-1}).

The absence of O_2 enhancement of the reduction of NO at 500°C with stoichiometric NH_3 for intermediate NO levels and with both stoichiometric and excess NH_3 for very low NO levels is not immediately obvious. Since the NH_3-O_2 reaction should predominate at 500°C, the observed effect may be related to a balance between the reduction of inlet NO and the formation of NO from NH_3 oxidation.

The effect of water vapor on NO reduction in simulated flue gas was studied by varying the water vapor concentration to 18% (simulation of methane combustion). The preliminary test results given in Table 9 indicated that NO conversion was not appreciably changed between water vapor concentrations of 0 to 10%. Additional data presented in Table 10 and Figure 22 show that conversion of 855 to 1030 ppm NO was not affected by variable H_2O concentrations with either stoichiometric or excess NH_3.

The preliminary observation that CO_2 had only a slight effect on NO conversion (Table 9) was confirmed by the results of additional tests shown in Table 11 and Figure 23. The effect on NO conversion over practical CO_2 concentrations between 8 to 16% can be considered to be negligible.

A study of the effect of residence time (or space velocity) was made. Typical data are shown in Figure 24 along with representations of data taken at 20,000 hr^{-1} in earlier studies. It can be seen that conversion of NO is not significantly affected by reactor volume at a constant space velocity (10,000 hr^{-1}) indicating negligible bulk diffusion effects at that space velocity. An increase in space velocity (decrease in residence time) causes a definite decrease in conversion as expected.

The fact that CO_2 and H_2O had negligible effects on the rate of reduction of NO eliminated the necessity for incorporating concentration terms for these species in the rate expression. The slight increase in conversion of NO with increasing O_2 concentration above 0.5% indicated that a power-law reaction order for O_2 would be finite but quite small.

The parametric data were correlated for individual concentrations of O_2 by the method of minimization of the sum of squares of the difference between experimental and calculated reaction rate constants, as described in Section 4. Initial data reduction was based on the global rate for the test system employed. In this initial analysis, bulk and internal diffusion effects are not considered. A least square fit of the logarithm of the rate at various O_2 partial pressures versus the logarithm of O_2 partial pressure was then performed for the individual O_2 concentrations. The rates used in this regression were calculated from best-fit reaction-rate parameters (E, activation energy, A, pre-exponential factor, n, reaction order for NO and m, reaction order for NH_3) determined for sets of data at constant O_2 concentrations.

The apparent reaction order for O_2 determined by the above technique was 0.05. A rate expression of the form,

TABLE 10. EFFECT OF WATER VAPOR CONCENTRATION ON REDUCTION OF NO ON V_2O_5 IN SIMULATED FLUE GAS

Inlet Gas Composition					Outlet Gas Comp.			Temp.	Conversion of	
NO_x (ppm)	NH_3 (ppm)	CO_2 (%)	H_2O (%)	O_2 (%)	NO_x (ppm)	NH_3 (ppm)	N_2O (ppm)	(°C)	NO (%)	NH_3 (%)
855	664	12	0	3	290	0	0	400	66	100
"	"	"	1	"	300	0	0	"	65	"
915	"	"	3	"	285	0	0	"	69	"
950	653	"	5	"	305	0	0	"	68	"
990	"	"	10	"	350	0	0	"	65	"
1050	"	"	18	"	370	0	0	"	"	"
915	1433	12	0	3	150	256	0	400	83	82
"	"	"	1	"	160	"	0	"	32	"
970	"	"	3	"	164	490	0	"	83	66
990	"	"	5	"	"	559	0	"	"	61
1030	"	"	10	"	170	629	0	"	"	56
1050	"	"	18	"	"	676	0	"	84	53

---

s.v. = 20,000 hr^{-1}

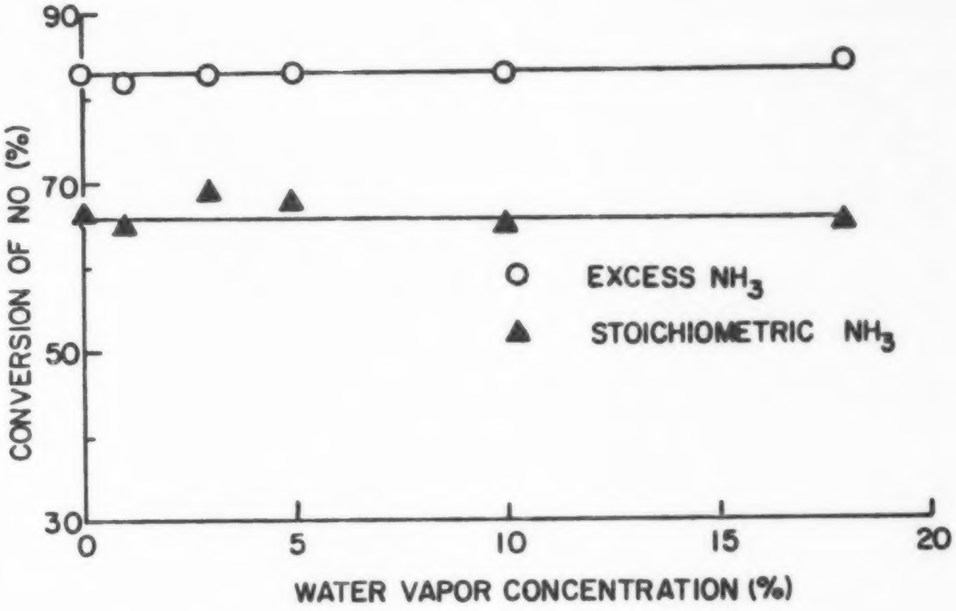

Figure 22. Effect of water vapor on NO reduction in simulated flue gas at 400°C on V_2O_5. 855-1030 ppm NO, 12% CO_2, 3% O_2. (s.v.= 20,000 hr^{-1}).

TABLE 11. EFFECT OF CO_2 CONCENTRATION ON CONVERSION OF NO ON V_2O_5

Inlet Gas Composition					Outlet Gas Comp.			Temp.	Conversion of	
NO_x (ppm)	NH_3 (ppm)	CO_2 (%)	H_2O (%)	O_2 (%)	NO_x (ppm)	NH_3 (ppm)	N_2O (ppm)	(° C)	NO (%)	NH_3 (%)
915	1468	0	5	3	100	513	0	400	89	65
995	"	1	"	"	125	"	0	"	87	"
"	"	3	"	"	130	524	0	"	86	64
"	"	5	"	"	140	559	0	"	"	62
990	"	12	"	"	160	489	0	"	84	67
995	"	14	"	"	170	559	0	"	83	62
"	"	20	"	"	170	524	0	"	"	64
965	746	0	5	3	305	105	0	400	68	86
"	"	1	"	"	"	58	0	"	"	92
1000	"	3	"	"	340	70	0	"	66	91
990	"	5	"	"	"	"	0	"	"	"
"	"	12	"	"	355	82	0	"	64	89
"	"	14	"	"	365	70	0	"	63	91
995	"	20	"	"	360	"	0	"	64	"

---

s.v. = 20,000 hr^{-1}

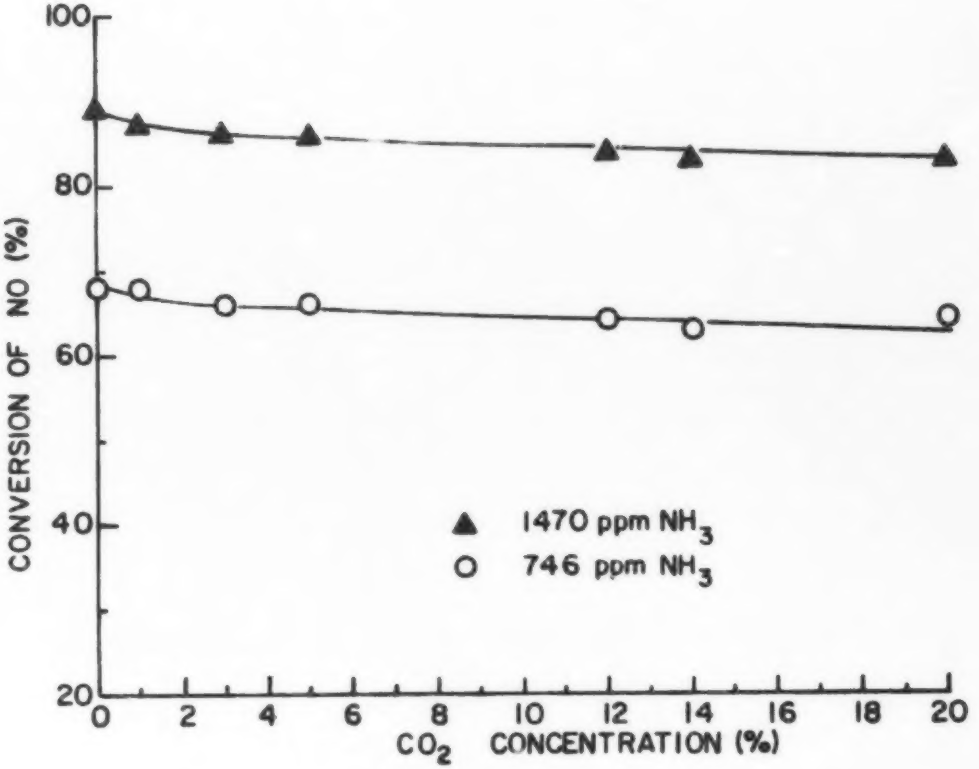

Figure 23. Effect of CO_2 on NO conversion in simulated flue gas at 400°C on V_2O_5. 915-995 ppm NO, 3% O_2, 5% H_2O. (s.v.= 20,000 hr^{-1}).

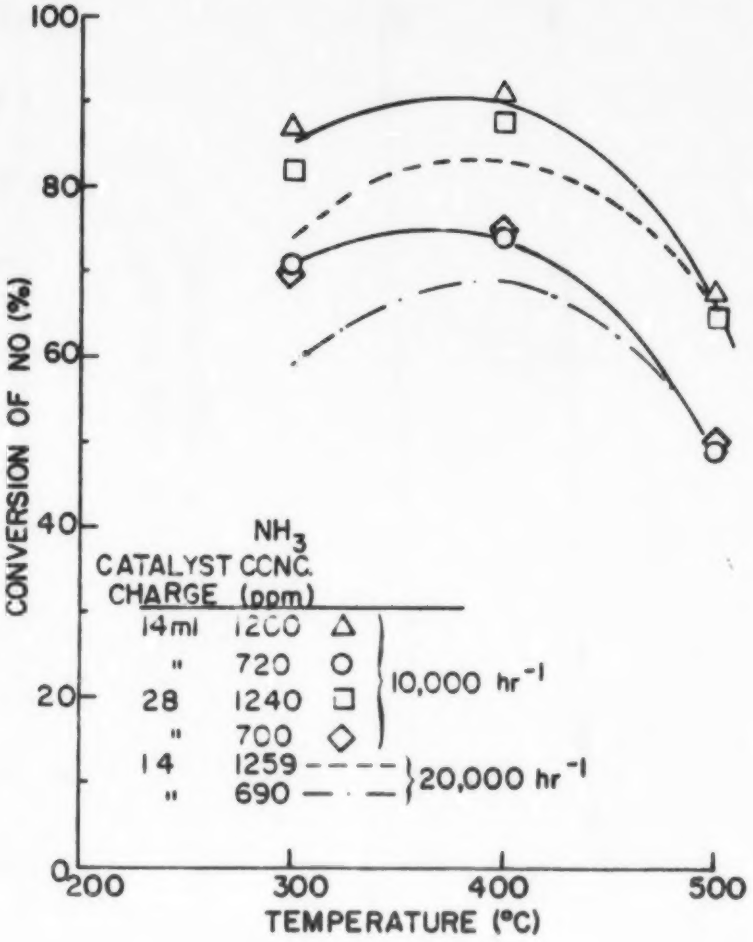

Figure 24. Effect of space velocity on conversion of NO in simulated flue gas on V_2O_5. 980 ppm NO, 12% CO_2, 3% O_2, 5% H_2O.

$$\text{rate} = Ae^{-E/RT} p_{NO}^{n} p_{NH_3}^{m} p_{O_2}^{0.05}$$

was then used.

A second iteration of the procedure for the determination of A, E, n and m was performed to refine the preliminary values. The new parameters thus obtained were used in a second computer program which determines the conversion of NO from the assumed rate expression for the experimental inlet concentrations and temperatures (see Section 4). As before, interparticle and intraparticle diffusion resistances were not considered in the determination of the global rate expression. The calculated and experimental values for conversion were then compared and several additional adjustments in the reaction rate parameters were made until a minimum in the differences between calculated and experimental conversion was obtained.

The final global rate expression which provides the best agreement between experimental and calculated conversion values for $T \leq 400^\circ C$ is

$$\text{rate} = 14.8\, e^{-3650/RT} p_{NO}^{0.5} p_{NH_3}^{0.4} p_{O_2}^{0.05} \text{(moles NO/gm catalyst - hr)} \qquad \text{(A)}$$

It should be emphasized that the above power law rate expression is only an empirical representation of the experimental data and does not imply any actual reaction mechanism.

The global rate expression (A) was then used as a first approximation of the intrinsic rate expression (which describes the rate of the surface reaction) in the iterative calculation of the conversion of NO in which the effect of external and internal mass and heat transfer was considered. The step-by-step procedure involved the determination of the effectiveness factor for reactor length increments. The individual kinetic parameters (A, E, m, n) were adjusted until the best agreement between experimental and calculated percent conversion of NO for all runs was achieved. The intrinsic rate expression that best describes the data up to 400°C is,

$$\text{rate} = 2.05 \times 10^{3}\, e^{-9650/RT} p_{NO}^{0.30} p_{NH_3}^{0.22} p_{O_2}^{0.05} \text{(moles NO/g cat-hr)} \qquad \text{(B)}$$

Equation (B) predicts conversion of NO_x well, as shown in Tables 12a and 12b , for space velocities of 20,000 hr^{-1} and 10,000 hr^{-1}, respectively. The differences in the activation energy and the reaction orders between Equations (A) and (B), and the low effectiveness factor values indicate substantial pore diffusion effects.

For temperatures above 400°C, two approaches were used to establish a workable global rate expression. The simplest technique was to fit directly the experimental data for NO conversion to a power law rate expression. The resulting expression which provided the best fit of all data for $T \geq 400^\circ C$ is:

TABLE 12a. EXPERIMENTAL AND CALCULATED CONVERSION OF NO ON V_2O_5-Al_2O_3 BASED ON INTRINSIC RATE EXPRESSION (20,000 hr^{-1}, Temperature $\leq 400^oC$)

Temp. (^oC)	Inlet Partial Pressures P^o_{NO} (atm x 10^6)	$P^o_{NH_3}$ (atm x 10^6)	$P^o_{O_2}$ (atm x 10^2)	Conversion Exptl. (%)	Calculated (%)	η, Effectiveness Factor
205	285	315	3	39	40	0.14
200	275	359	3	31	40	0.14
200	280	365	3	32	40	0.14
200	770	921	3	27	33	0.18
200	770	1189	3	27	34	0.17
200	985	1259	3	27	32	0.18
200	980	957	3	28	31	0.19
200	1005	1485	3	27	33	0.18
200	1475	1002	3	26	28	0.22
200	1410	1457	3	26	29	0.21
200	1485	2400	3	28	31	0.19
200	1485	1981	3	27	30	0.20
200	1460	785	3	25	24	0.18
305	285	315	3	77	71	0.05
300	280	245	3	63	68	0.05
305	785	769	3	70	62	0.06
300	805	501	3	61	56	0.07
300	770	1189	3	72	63	0.06
300	770	921	3	70	62	0.06
300	980	967	3	65	59	0.07
300	995	1468	3	65	59	0.07
300	1460	967	3	56	52	0.08
300	1410	1457	3	67	57	0.07
300	1485	2400	3	62	58	0.07
300	1475	1981	3	61	58	0.07
300	510	734	3	71	66	0.05
395	770	1189	3	87	84	0.02
400	785	769	3	80	83	0.03
400	770	921	3	84	84	0.02
400	770	435	3	74	69	0.03
400	985	1259	3	84	83	0.03
400	980	967	3	79	81	0.03
400	850	1165	3	82	84	0.02
400	990	1433	3	83	83	0.02
400	990	1468	3	84	83	0.02
395	1420	967	3	73	73	0.04
400	1410	1457	3	83	79	0.03
400	1485	2400	3	78	80	0.03

(continued)

TABLE 12 a (cont.)

Temp.	Inlet Partial Pressures			Conversion		η, Effectiveness Factor
(°C)	P^o_{NO} (atm x 10^6)	$P^o_{NH_3}$ (atm x 10^6)	$P^o_{O_2}$ (atm x 10^2)	Exptl. (%)	Calculated (%)	
400	1475	1981	3	85	80	0.03
300	770	1095	3	79	84	0.02
300	510	734	5	73	67	0.05
300	510	734	1	68	65	0.05
300	565	734	0.5	65	63	0.06
300	500	385	3	63	62	0.06
300	490	385	5	63	63	0.06
300	265	408	3	66	72	0.04
300	570	385	1	58	58	0.07
300	595	385	0.5	55	57	0.07
300	865	1130	0.5	65	60	0.06
300	790	580	3	62	58	0.07
300	850	580	1	55	55	0.08
300	870	580	0.5	52	54	0.08
300	795	580	5	60	59	0.07
300	990	1491	0.5	62	59	0.06
300	1000	652	3	55	55	0.08
300	980	652	5	59	56	0.07
300	1020	641	1	53	53	0.08
300	1075	641	0.5	51	51	0.08
300	1050	1445	1	63	59	0.06
395	285	315	3	86	89	0.02
400	760	1095	5	80	85	0.02
400	800	1107	3	80	84	0.02
400	870	1107	1	79	83	0.03
400	890	1107	0.5	77	81	0.03
395	995	1468	3	89	83	0.03
400	1030	1480	3	91	83	0.02
400	990	1442	3	86	83	0.02
400	990	1442	5	89	84	0.02
400	990	1442	1	83	82	0.03
400	990	1442	0.5	80	81	0.03
400	1060	676	0.5	71	73	0.04
200	285	256	3	32	38	0.15
200	805	536	3	22	30	0.20
200	785	769	3	26	32	0.18
200	770	455	3	24	29	0.19
200	980	664	3	29	29	0.21
200	980	583	3	28	28	0.20

TABLE 12b. EXPERIMENTAL AND CALCULATED CONVERSION OF NO ON V_2O_5-Al_2O_3 BASED ON INTRINSIC RATE EXPRESSION (10,000 hr^{-1}, Temperature $\leq$ 400°C)

Temp.	Inlet Partial Pressures			Conversion		η, Effectiveness Factor
(°C)	P^o_{NO} (atm x 10^6)	$P^o_{NH_3}$ (atm x 10^6)	$O^o_{NH_3}$ (atm x 10^2)	Exptl. (%)	Calculated (%)	
300	960	722	3	71	71	0.09
400	970	1189	3	91	94	0.02
300	970	1189	3	87	75	0.08
300	980	699	3	71	71	0.10
300	970	1239	3	82	75	0.08
400	970	1239	3	87	84	0.03
400	1020	1445	3	90	94	0.02
400	1000	932	3	86	93	0.03

$$\text{rate} = 0.30\ e^{8050/RT}\ p_{NO}^{0.9}\ p_{NH_3}^{0.5}\ p_{O_2}^{0.2}\ \text{(moles NO/gm catalyst-hr)} \qquad \text{(C)}$$

The negative activation energy predicts decreasing rates of NO conversion with increasing temperature, as observed experimentally. Table 13 shows reasonable agreement between experimental and calculated conversion of NO. The largest discrepencies occurred at 500°C with excess NH_3.

For the conditions to which Equation (C) applies, the apparent, net conversion of NO decreases with increasing temperature because (a) NH_3 is being depleted due to reaction with O_2 and (b) under some conditions, NO is the product of NH_3 oxidation with O_2. It can be shown that in the absence of such competing reactions, the actual rate of reduction of NO does not pass through a maximum over the temperature range of this study.

A second method examined for correlating the kinetic data above 400°C used Equation (A) in conjunction with an expression for NH_3 reaction due to decomposition and oxidation by O_2 as measured under NO-free conditions. As an approximation, it was assumed that the reactions were consecutive and the appropriate NH_3 partial pressure for Equation (1) was that remaining after the latter reactions. This study was not successful. It is quite apparent that the individual reactions are not consecutive as assumed. In fact, it has been shown (5-1) that even in the ascending-temperature region, for which an adequate empirical rate expression was obtained, the reactions of NO, NH_3 and O_2 are interrelated in a complex manner.

An analysis of the experimental kinetic data above 400°C considering external and internal diffusion was not attempted.

5.2.1.2. Fe-Cr Catalyst

The selected 10% iron-chromium oxide catalyst with an Fe/Cr ratio of 9/1 was submitted to a parametric test program following the general procedure used for V_2O_5.

Preliminary tests were concerned primarily with establishing the effects of NO and NH_3 concentration on conversion of NO. Table 14 summarizes typical data for temperatures from 200 to 490°C with NO varying from about 320 to 1400 ppm and NH_3 varying in amount slightly below stoichiometric to a 3/1 ratio with respect to NO. The standard flue gas composition (12% CO_2, 3% O_2, 5% H_2O in N_2) was used at 20,000 hr^{-1} space velocity. Nitrous oxide was produced only with large concentrations of NO at 490°C. The data indicate that conversion of NO is a function of the NH_3/NO ratio as observed with the V_2O_5 catalyst. Figure 25 shows this dependency for temperatures up to 395°C.

As with the V_2O_5 catalyst, conversion of NO did not vary above NH_3/NO ratios of about 1.1 (0.67 NH_3/NO is stoichiometric for conversion of NO to N_2). For NH_3/NO < 1, a change in the reactant ratio has a significant effect on the conversion of NO,

TABLE 13. EXPERIMENTAL AND CALCULATED CONVERSION OF NO ON V_2O_5-Al_2O_3 BASED ON GLOBAL RATE EXPRESSION (Temp. $\geq 400^{\circ}C$)

Temp. (°C)	p°_{NO} (atm x 10^6)	$p^{\circ}_{NH_3}$ (atm x 10^6)	$p^{\circ}_{O_2}$ (atm x 10^2)	Conversion Exptl. (%)	Conversion Calculated (%)
400	1485	2400	3	88	90
400	1410	1457	3	83	82
395	995	1468	3	89	86
400	980	583	3	59	64
400	805	501	3	61	63
400	770	921	3	84	78
400	990	1442	0.5	80	75
400	1060	676	0.5	63	57
400	890	1110	0.5	77	70
400	970	688	5	71	71
400	990	1442	5	89	87
400	1020	688	1	66	61
400	990	1442	1	83	79
400	870	1110	1	79	75
395	1475	1980	3	85	88
395	1460	967	3	63	72
400	1030	1480	3	91	85
395	995	1468	3	89	86
400	990	1440	3	86	85
400	985	1259	3	84	83
400	980	967	3	79	77
400	985	690	3	69	68
400	950	655	3	68	67
400	985	688	3	69	68
400	975	524	3	62	61
400	950	1165	3	82	81
400	785	769	3	80	74
400	770	921	3	84	78
490	275	350	3	47	44
490	280	245	3	50	38
500	285	186	3	35	32
495	805	501	3	45	42
500	785	769	3	64	50
500	770	1189	3	73	59
490	975	524	3	45	42
495	995	1468	3	72	63
480	980	967	3	61	54
485	985	1259	3	69	60

(continued)

TABLE 13. (cont.)

Temp. (°C)	p°_{NO} (atm x 10^6)	$p^\circ_{NH_3}$ (atm x 10^6)	$p^\circ_{O_2}$ (atm x 10^2)	Conversion Exptl. (%)	Conversion Calculated (%)
495	980	583	3	45	44
495	750	548	3	50	45
495	505	781	3	48	55
495	275	291	3	35	40
440	265	180	3	57	52
500	520	380	0.5	27	31
500	520	352	1.0	29	33
500	520	769	0.5	39	42
495	860	1105	1.0	56	51
495	800	548	1.0	36	38
495	815	548	0.5	33	35

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s.v. = 20,000 hr^{-1}. Simulated flue gas contains also 12% CO_2, 5% H_2O in N_2.

TABLE 14. PARAMETRIC STUDIES TEST DATA - Fe-Cr CATALYST IN SIMULATED FLUE GAS.

Inlet Gas Comp.		Outlet Gas Comp.				Conv. of
NO_x (ppm)	NH_3 (ppm)	NO_x (ppm)	NH_3 (ppm)	N_2O (ppm)	Temp. (°C)	NO_x (%)
950	746	920	676	0	200	3
940	957	890	862	0	200	5
955	1305	940	1200	0	200	12
950	1859	815	1773	0	200	14
805	559	765	524	0	200	5
815	670	770	629	0	200	6
805	897	755	823	0	200	6
740	1340	590	1142	0	200	20
420	513	400	466	0	200	5
465	559	430	478	0	200	7
450	722	420	664	0	200	7
465	1165	385	1085	0	200	17
345	489	325	466	0	200	6
335	676	315	629	0	200	6
340	1026	310	956	0	200	9
315	475	205	315	0	300	35
325	618	195	443	0	300	40
325	862	190	676	0	300	42
505	536	280	291	0	300	45
480	862	300	618	0	300	38
465	886	255	629	0	300	45
470	618	280	396	0	300	40
460	1084	255	850	0	300	45
915	723	640	147	0	295	30
915	1026	630	850	0	295	31
745	490	430	233	0	300	42
800	653	450	385	0	300	44
745	1002	410	653	0	300	45
760	1328	440	962	0	300	42
1325	724	920	375	0	300	31
1375	897	915	501	0	300	33
1375	1375	900	897	0	300	34
1325	2293	860	1643	0	300	35

(continued)

TABLE 14. (cont.)

Inlet Gas Comp.		Outlet Gas Comp.				Conv. of
NO_x (ppm)	NH_3 (ppm)	NO_x (ppm)	NH_3 (ppm)	N_2O (ppm)	Temp. (°C)	NO_x (%)
490	678	245	290	0	390	50
535	1077	215	642	0	390	60
545	1573	210	1040	0	390	61
1000	1574	305	546	0	440	70
1000	2905	295	1420	0	440	71
1010	2905	270	1307	0	480	73
490	520	165	121	0	440	66
490	884	155	278	0	440	68
480	1150	155	448	0	440	68
480	1450	140	932	0	440	71
490	884	155	278	0	440	68
480	1105	140	448	0	440	71
480	1450	155	932	0	440	68
490	520	165	121	0	440	66
490	460	215	60	0	480	56
515	775	185	106	0	480	64
515	1132	185	206	0	495	64
515	1524	175	355	0	490	66
755	569	370	48	0	490	51
770	775	300	97	0	490	61
765	1524	215	290	0	490	72
765	2290	215	533	0	490	72
1010	557	505	0	0	445	50
985	920	285	133	0	445	71
955	1162	255	254	0	445	73
965	1474	240	399	0	445	75
990	800	390	89	0	490	61
1040	920	340	121	0	490	67
900	1215	235	166	0	490	74
990	1501	230	266	0	490	77

(continued)

TABLE 14 (cont.)

Inlet Gas Comp.		Outlet Gas Comp.				
NO_x (ppm)	NH_3 (ppm)	NO_x (ppm)	NH_3 (ppm)	N_2O (ppm)	Temp. (°C)	Conv. of NO_x (%)
1425	690	830	0	0	485	42
1435	1132	685	73	65	490	52
1415	1573	490	254	80	490	65
1435	2397	435	440	75	490	70
870	1305	260	400	0	400	70
770	569	285	48	0	400	63
795	762	275	169	0	400	65
785	1015	270	363	0	400	66
785	1307	225	678	0	400	71
890	1015	295	339	0	400	67
520	460	220	121	0	400	58
515	544	215	230	0	400	58
520	859	205	470	0	400	61
560	1260	190	654	0	400	66
475	859	185	470	0	400	61
1300	508	1020	44	0	400	22
1400	508	1070	44	0	400	24
1345	820	740	157	0	400	45
1290	1130	605	277	0	400	53
1335	1586	450	375	0	440	66
1335	1586	390	12	0	485	71
780	763	235	204	0	440	70
805	1114	240	350	0	440	70
770	1935	205	859	0	440	73
390	524	165	237	0	360	58
405	792	160	490	0	360	60
395	1189	155	932	0	360	61
405	1573	165	1107	0	360	59
555	629	225	28	0	460	59
565	758	225	40	0	360	60
540	1141	210	718	0	360	61
555	1573	205	1038	0	360	63

(continued)

TABLE 14. (cont.)

Inlet Gas Comp.		Outlet Gas Comp.				
NO_x (ppm)	NH_3 (ppm)	NO_x (ppm)	NH_3 (ppm)	N_2O (ppm)	Temp. (°C)	Conv. of NO_x (%)
1035	522	515	77	0	360	50
1030	788	390	163	0	360	62
1060	1119	380	454	0	360	64
1080	1608	370	839	0	360	66
1295	1130	500	443	0	360	61
1340	1608	480	769	0	360	64
965	533	490	73	0	395	49
965	932	300	194	0	395	69
965	1065	290	327	0	395	70
965	1465	270	617	0	395	72

20,000 hr^{-1} (STP) space velocity

12% CO_2, 5% H_2O, 3% O_2 in N_2 carrier gas present.

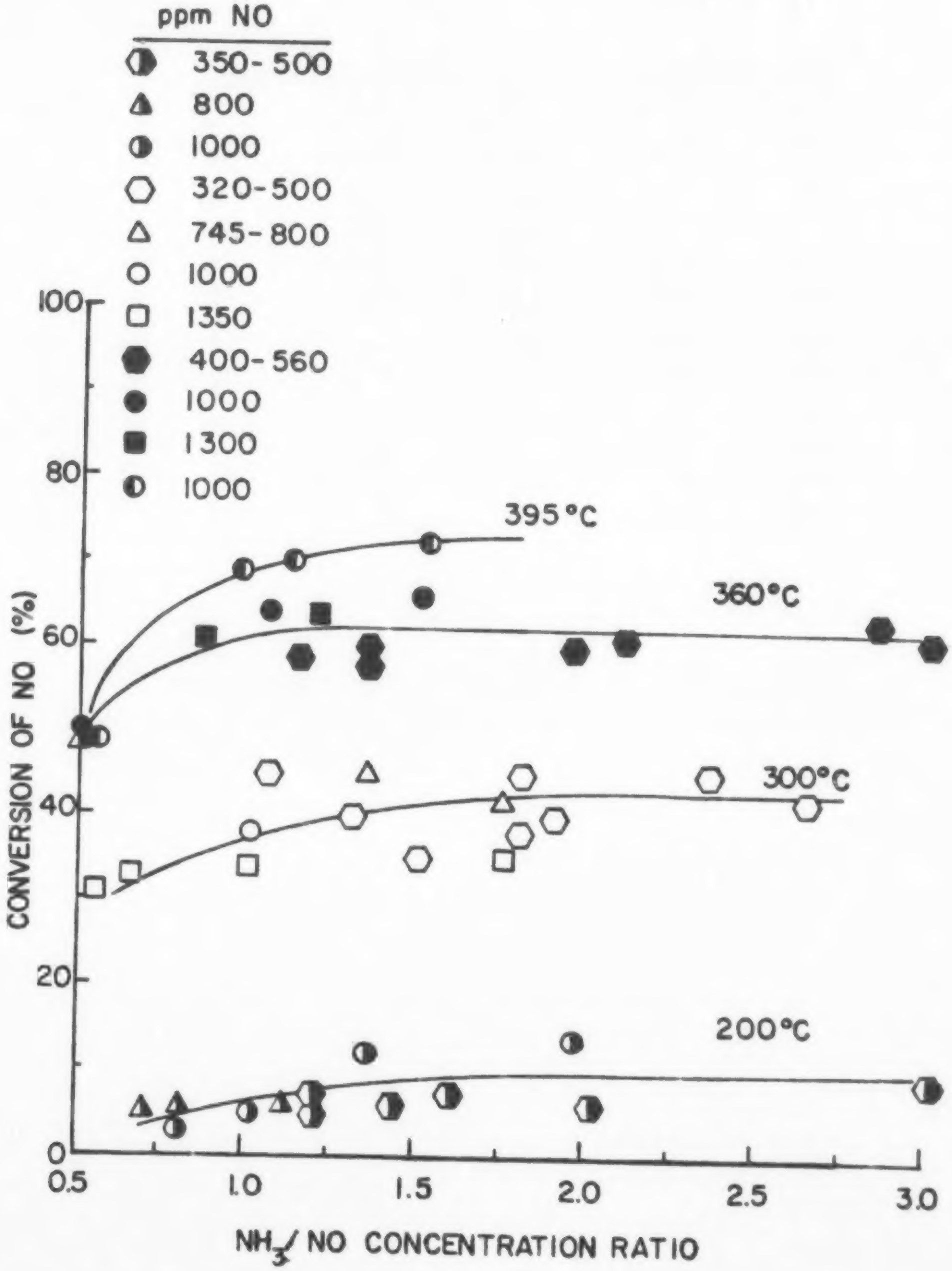

Figure 25. Conversion of NO in simulated flue gas on Fe-Cr catalyst. (s.v.= 20,000 hr-1).

and there is a trend toward increasing dependency on the reactant ratio with increasing temperature. This result was also observed with the V_2O_5 catalyst. For temperatures of 390°C and higher, Figure 26 shows that the conversion of NO is not dependent on the individual concentrations of NO and NH_3 but dependent on the NH_3/NO ratio. The lower and upper curves are for 400 and 440°C, respectively. The data indicate that a broad maximum in conversion on the Fe-Cr catalyst occurred at 440°C and a sharp decrease at temperatures above 400°C was not observed as was observed for the V_2O_5 catalyst. Figure 26 also shows that conversion at 490°C (half-filled points) is equivalent to that at 400°C for low NH_3/NO ratios and to that at 440°C for higher NH_3/NO ratios.

The broad maximum in conversion for the Fe-Cr catalyst offers the operational advantage that temperature regulation of the reactor will not be as strict as for the V_2O_5 catalyst. A single rate expression for design use up to 450°C should be applicable. For instance, nominal operation at 425°C could tolerate a ± 25° temperature variation with only a 2% change in conversion.

The effects of H_2O and CO_2 on NO conversion were determined by varying the H_2O and CO_2 concentrations about the nominal, simulated flue gas values. Carbon dioxide concentration was varied from 0 to 20% (nominal = 12%) and water vapor was varied from 0 to 18% (nominal = 5%). These effects were studied at 400 and 450°C with both stoichiometric and excess NH_3 for the CO_2 tests and with excess NH_3 for the H_2O tests.

Table 15 presents typical data for the parametric study. Figures 27 and 28 show that neither CO_2 nor H_2O have any appreciable effect on the conversion of NO_x. As with the V_2O_5 catalyst, such insensitivity to either constituent significantly simplifies the development of the rate expression.

Oxygen concentration was varied from 0 to 5% to determine the extent of the enhancement effect. Data were obtained for NO concentrations of about 750 and 950 ppm with stoichiometric and excess NH_3 at temperatures from 300 to 450°C. Table 16 shows some typical data.

Figure 29 shows that with stoichiometric or excess NH_3 and at all temperatures studied, the presence of a small amount of O_2 produces substantial enhancement of the conversion of NO. Above 0.5% an increase in O_2 affects conversion only slightly. By assuming that only O_2 levels above 0.5% will be considered for design use (low excess air firing cannot realistically be pushed to this low limit), the data indicate a positive, near-zero reaction order for O_2. With V_2O_5, a low reaction order (0.05) was also found. The nearly parallel NO conversion - O_2 concentration curves above 0.5% O_2 indicate that the reaction order for O_2 will be essentially independent of temperature and NO/NH_3 ratio.

The effect of space velocity on conversion of NO was assessed in a series of

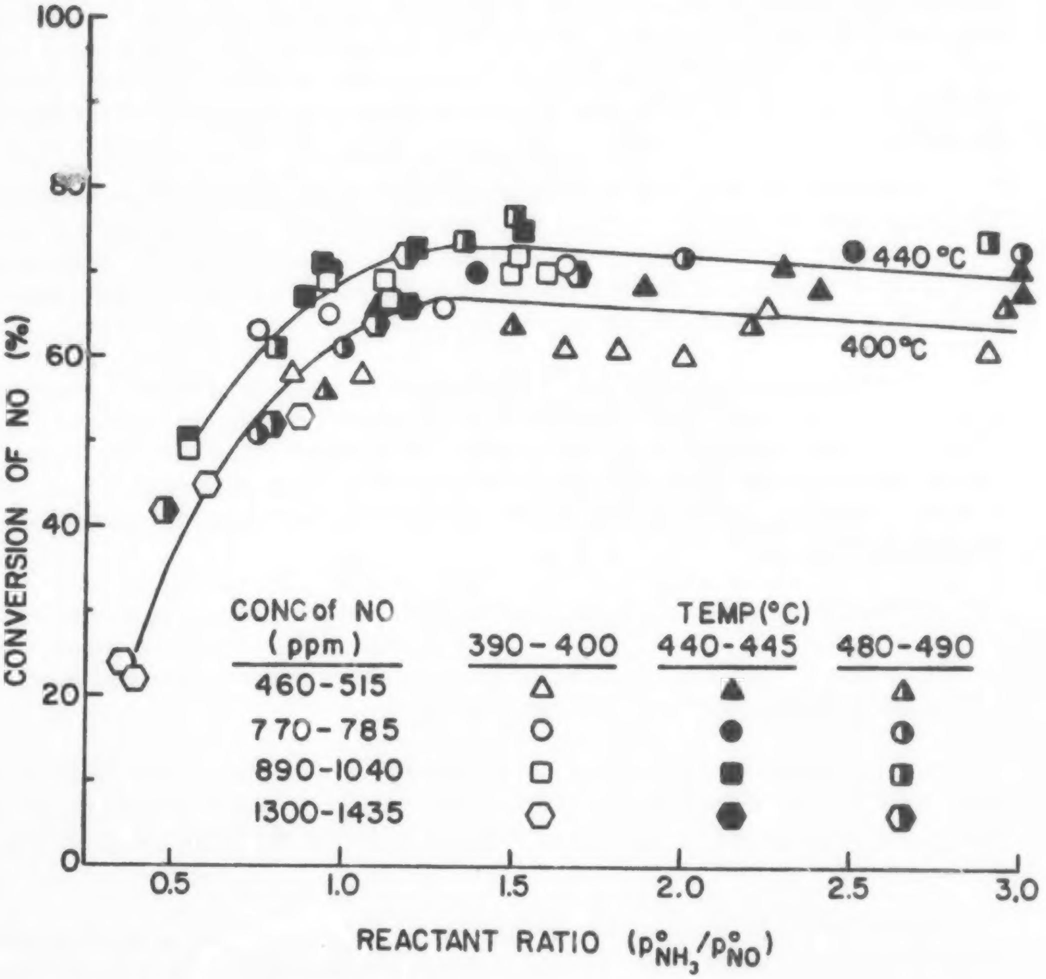

Figure 26. Conversion of NO over Fe-Cr catalyst at temperatures above 390°C in simulated flue gas. (s.v.= 20,000 hr^{-1}).

TABLE 15. PARAMETRIC STUDIES TEST DATA - Fe-Cr CATALYST IN SIMULATED FLUE GAS -EFFECT OF WATER VAPOR AND CARBON DIOXIDE

Inlet Gas Composition					Outlet Gas Comp.				Conv. of
NO_x (ppm)	NH_3 (ppm)	CO_2 (%)	H_2O (%)	O_2 (%)	NO_x (ppm)	NH_3 (ppm)	N_2O (ppm)	Temp (°C)	NO (%)
1005	690	12	5	3	470	178	0	400	53
1090	690	20	5	3	510	142	0	400	53
1005	690	14	5	3	480	169	0	400	52
1070	707	5	5	3	480	169	0	400	55
1070	707	3	5	3	480	157	0	400	55
1060	707	0	5	3	480	145	0	400	55
1000	671	12	5	3	450	85	0	445	55
1075	717	20	5	3	460	73	0	445	57
1000	738	14	5	3	450	85	0	445	55
1045	717	5	5	3	480	83	0	445	54
1040	763	3	5	3	480	73	0	445	54
1040	749	0	5	3	475	73	0	445	54
1055	1185	12	5	3	390	472	0	390	63
1050	1186	14	5	3	395	460	0	390	62
1090	1186	10	5	3	390	460	0	390	64
1090	1186	5	5	3	385	484	0	390	65
1095	1186	0	5	3	390	506	0	390	64
1020	1198	12	5	3	290	280	0	445	72
1030	1198	14	5	3	305	293	0	445	70
1060	1198	10	5	3	300	266	0	445	72
1095	1185	5	5	3	315	266	0	445	71
1060	1185	0	5	3	315	320	0	445	70
1035	1198	12	18	3	388	520	0	400	63
1070	1198	12	10	3	398	508	0	400	63
1050	1186	12	5	3	365	484	0	400	65
1080	1162	12	3	3	382	460	0	400	65
1105	1162	12	0	3	373	351	0	400	66
1147	1235	12	18	3	353	387	0	450	69
1127	1154	12	10	3	331	363	0	450	71
1040	1198	12	5	3	300	303	0	450	71
1100	1186	12	3	3	285	278	0	450	74
1094	1162	12	0	3	326	182	0	450	70

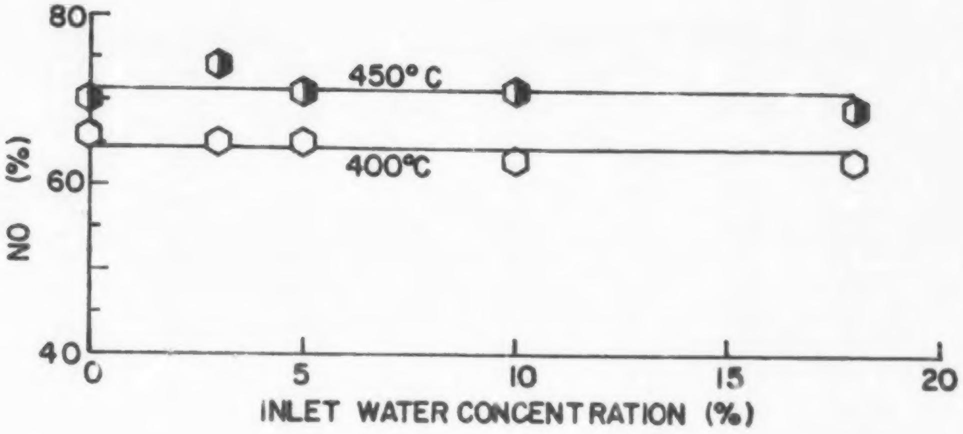

Figure 27. Effect of water vapor concentration on conversion of NO in simulated flue gas over Fe-Cr catalyst (12% CO_2, 3% O_2, 1000 ppm NO, 1200 ppm NH_3 in N_2). (s.v.= 20,000 hr^{-1}).

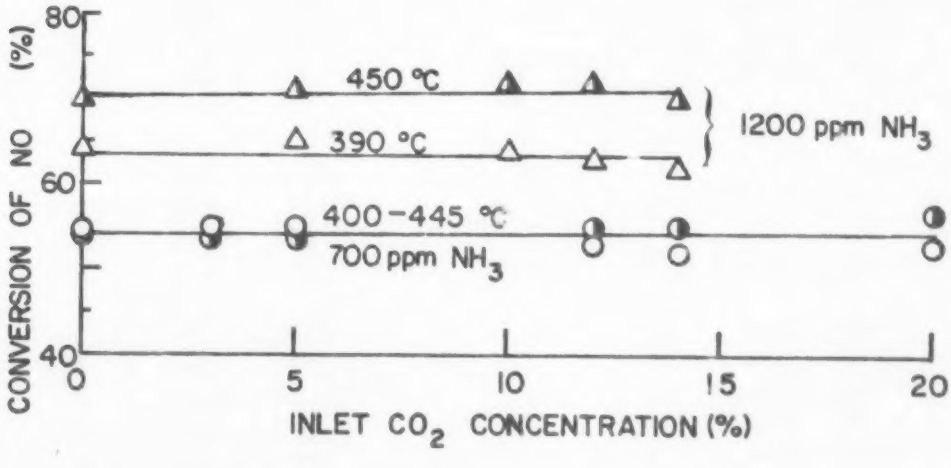

Figure 28. Effect of CO_2 concentration on conversion of NO in simulated flue gas over Fe-Cr catalyst (5% H_2O, 3% O_2, 1000 ppm NO in N_2, NH_3 as specified). (s.v. = 20,000 hr^{-1}).

TABLE 16. EFFECT OF OXYGEN CONCENTRATION ON NO REDUCTION WITH NH_3 IN SIMULATED FLUE GAS -Fe-Cr CATALYST

Inlet Gas Comp.			Outlet Gas Comp.				Conv. of
NO_x (ppm)	NH_3 (ppm)	O_2 (%)	NO_x (ppm)	NH_3 (ppm)	N_2O (ppm)	Temp. (°C)	NO (%)
720	546	0	670	484	0	300	7
735	546	0.5	500	346	0	300	32
745	506	1	480	333	0	300	36
760	506	3	460	320	0	300	39
750	506	5	435	266	0	300	42
720	866	0	650	759	0	305	10
735	852	0.5	475	626	0	305	35
740	923	1.0	465	618	0	305	37
740	852	3.0	490	545	0	305	34
950	796	0	875	666	0	305	8
940	799	0.5	680	472	0	305	28
960	799	1.0	640	436	0	305	33
975	764	3.0	590	417	0	305	39
1010	787	5.0	600	426	0	305	41
940	1227	0	905	1162	0	305	4
1010	1259	0.5	715	959	0	305	29
1015	1283	1.0	680	908	0	305	33
1030	1283	3.0	645	932	0	305	37
1065	1283	5.0	630	847	0	305	41
725	908	0	615	820	0	400	15
720	908	0.5	305	411	0	400	58
760	905	1.0	295	436	0	400	61
760	905	3.0	265	389	0	400	65
800	905	5.0	265	387	0	400	67
920	750	0	820	493	0	395	13
950	750	0.5	475	181	0	395	50
960	750	1.0	455	160	0	395	53
990	706	3.0	445	133	0	395	55
1005	653	5.0	440	121	0	395	56

(continued)

TABLE 16 (cont.)

Inlet Gas Composition			Outlet Gas Composition				Conv. of
NO_x (ppm)	NH_3 (ppm)	O_2 (%)	NO_x (ppm)	NH_3 (ppm)	N_2O (ppm)	Temp (° C)	NO_x (%)
955	750	1	870	690	0	205	9
980	750	3	840	633	0	205	14
990	750	5	860	640	0	205	13
990	557	1	925	480	0	205	7
1030	557	3	920	440	0	205	11
995	557	5	900	425	0	205	10
760	587	1	690	533	0	205	9
795	587	3	660	---	0	205	17
735	557	1	680	520	0	205	8
750	557	5	645	488	0	205	14
760	557	1	610	460	0	260	20
765	557	3	580	425	0	260	24
740	610	5	560	405	0	260	24
730	777	1	590	725	0	260	19
775	777	3	570	700	0	260	26
760	777	5	555	654	0	260	27
1050	777	3	825	700	0	260	21
970	845	1	800	702	0	260	18
965	845	3	750	665	0	260	22
990	845	5	740	615	0	260	25
1285	1065	3	1050	847	0	260	18
1390	1065	1	1145	896	0	260	17
1265	1065	5	990	812	0	260	22
1015	1065	3	820	---	0	260	19
1345	760	1	1135	599	0	255	16
1460	760	5	1140	533	0	255	22
925	830	1	755	738	0	255	18
960	830	5	730	605	0	255	24
780	448	1	615	424	0	240	21
710	448	5	525	344	0	250	26
735	1100	1	505	980	0	300	31
770	1053	5	485	738	0	300	37
1015	1100	1	695	799	0	300	31
1055	1100	5	640	741	0	300	39

(continued)

TABLE 16 (cont.)

Inlet Gas Composition			Outlet Gas Composition				
NO_x (ppm)	NH_3 (ppm)	O_2 (%)	NO_x (ppm)	NH_3 (ppm)	N_2O (ppm)	Temp (°C)	Conv. of NO_x (%)
740	714	1	360	363	0	355	51
760	714	5	315	278	0	355	58
955	810	1	515	424	0	355	46
960	810	5	435	322	0	355	54
945	1485	1	500	973	0	355	47
960	1485	5	410	884	0	355	57
1300	1529	1	735	826	0	355	43
1375	1529	5	630	666	0	355	54
1335	865	1	810	426	0	355	39
1405	865	5	750	280	0	355	47
740	1320	1	260	612	0	445	65
805	1318	5	225	400	0	445	72
755	639	1	280	213	0	450	63
790	639	5	240	120	0	450	70
1030	639	1	505	133	0	450	51
1060	639	5	460	100	0	450	57
1005	905	1	405	194	0	450	60
1095	905	5	340	206	0	450	69
1030	703	1	460	169	0	450	55
1105	726	5	410	107	0	450	63
990	1330	1	340	460	0	450	66
1050	1330	5	270	373	0	450	74
1020	1330	1	325	360	0	495	68
1040	1330	5	275	293	0	495	74
685	1235	1	205	426	0	485	70
780	1235	5	200	332	0	485	74
760	706	1	290	194	0	488	62
800	720	5	250	121	0	488	69

(continued)

TABLE 16. (cont.)

Inlet Gas Comp.			Outlet Gas Comp.				
NO_x (ppm)	NH_3 (ppm)	O_2 (%)	NO_x (ppm)	NH_3 (ppm)	N_2O (ppm)	Temp. (°C)	Conv. of NO (%)
920	1236	0	800	1017	0	395	13
920	1234	0.5	355	508	0	395	61
915	1122	1.0	345	488	0	395	62
960	1234	3.0	300	436	0	395	69
970	1162	5.0	315	392	0	395	68
730	932	0	525	639	0	450	28
750	918	0.5	275	266	0	450	63
765	905	1.0	260	242	0	450	66
765	905	3.0	235	182	0	450	69
795	905	5.0	230	226	0	450	71

20,000 hr^{-1} space velocity

5% H_2O, 12% CO_2 in N_2, other constituents as given above.

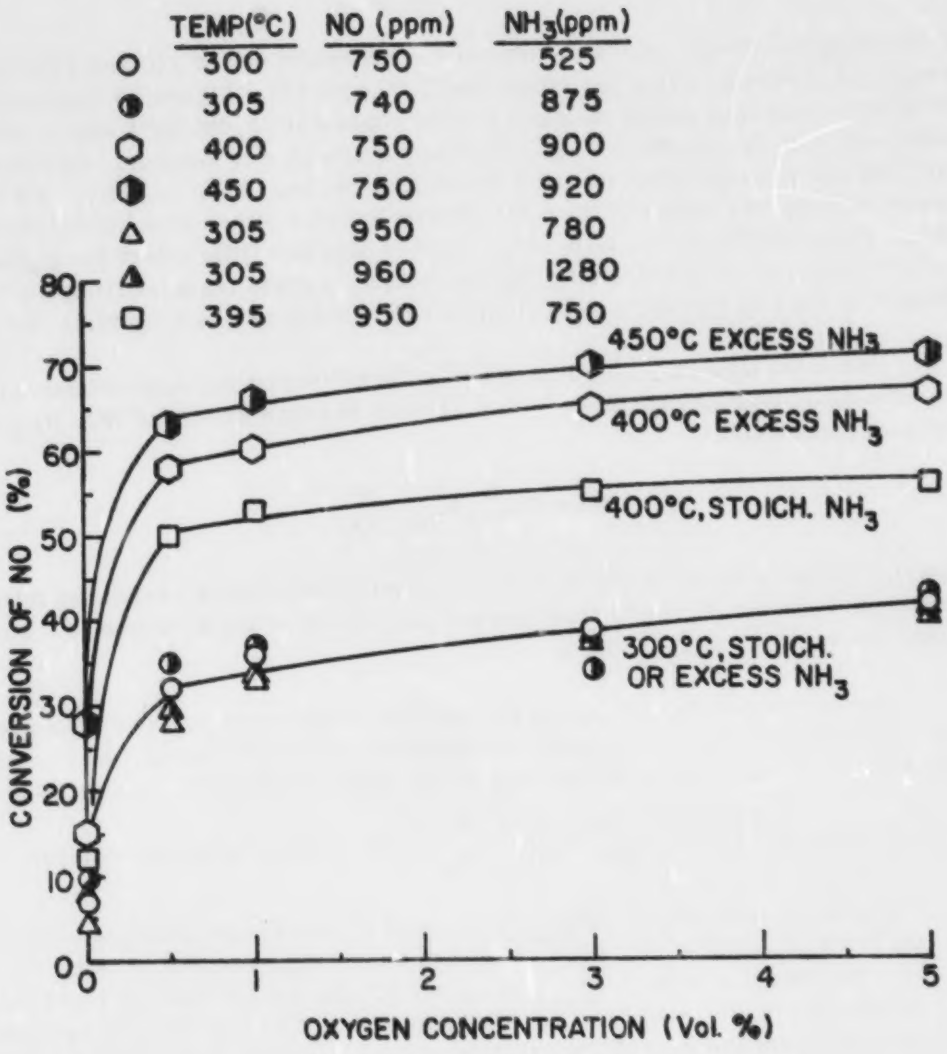

Figure 29. Effect of O_2 on conversion of NO with Fe-Cr catalyst in simulated flue gas (s. v.= 20,000 hr^{-1}).

tests using the 28-ml. reactor. Ammonia concentrations of 720 and 1200 ppm were employed in standard flue gas containing 1000 ppm NO. Figure 30 shows typical data along with representations of composite data taken at 20,000 hr^{-1} space velocity and additional data for 10,000 hr^{-1} (1/2 flow rate) in the 14 ml. reactor. The conversion maxima are seen to be broadened somewhat at the low space velocity. Above 400°C, space velocity has little effect on NO conversion with either stoichiometric or excess NH_3. Above 350°C with excess NH_3, reactor size has little effect for a given space velocity (10,000 hr^{-1}) indicating that resistance to mass transfer from the bulk gas stream to the catalyst surface is small for flow rates of that magnitude and larger.

The data were assembled in computer input format for determination of the specific reaction rate constants (k) for various reaction orders of NO, NH_3 and O_2 in the rate expression,

$$\text{rate} = k\, p_{NO}^{m}\, p_{NH_3}^{n}\, p_{O_2}^{q}$$

The sum of the squares of the difference between individually calculated rate constants and those determined from a least square regression of log k versus 1/T was determined for each set of m, n and q.

The reaction order values which resulted in the minimum sum of squares are given in the global rate expression (the apparent activation energy and pre-exponential factor were the slope and intercept of the linear regression, respectively).

$$\text{rate} = 32.4\, e^{-5400/RT}\, p_{NO}^{0.65}\, p_{NH_3}^{0.15}\, p_{O_2}^{0.15} \text{ (gm moles NO/hr-gm catalyst} \qquad \text{(D)}$$

The small influence of oxygen on the rate of conversion of NO is reflected in the low reaction order (0.15) which was, however, larger than that found for V_2O_5 (0.05). The reaction order of NO was higher for the Fe-Cr catalyst (0.65 compared to 0.5) and that of NH_3 was lower (0.15 compared to 0.4) than for the V_2O_5 catalyst. For both catalysts the sum of the reaction orders was 0.95 in the global rate expressions.

The very broad maximum in conversion of NO on the 9/1 Fe-Cr catalyst precludes separate rate expressions for the ascending and descending portions of the NO conversion-temperature curve. The single rate expression would be applicable to 450°C. Recommended operation is at 425°C, providing for a considerable temperature excursion (± 25°C).

The intrinsic rate expression best describing the kinetic data, developed in the iterative process described for the 9/1 Fe-Cr catalyst is:

$$\text{rate} = 3.25 \times 10^{3}\, e^{-10,860/RT}\, p_{NO}^{0.45}\, p_{NH_3}^{0.10}\, p_{O_2}^{0.15} \text{ (moles NO/gm catalyst/hr)} \qquad \text{(E)}$$

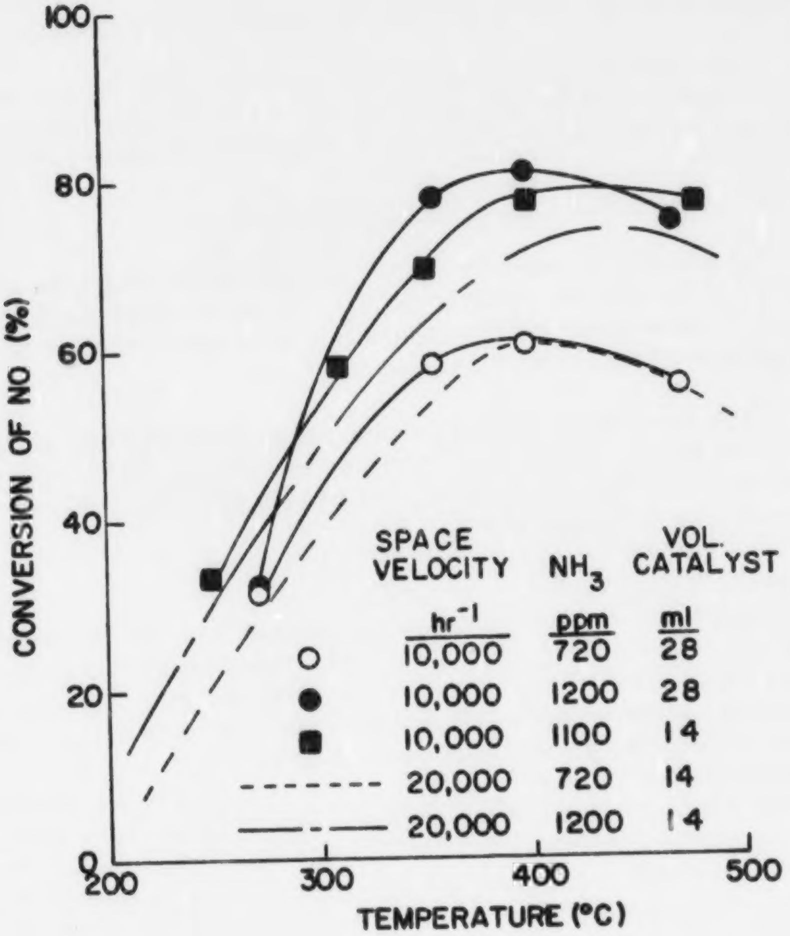

Figure 30. Effect of space velocity on conversion of NO in simulated flue gas on Fe-Cr. 1000 ppm NO, 12% CO_2, 3% O_2, 5% H_2.

Tables 17a and 17b list experimental and calculated conversion of NO based on Equation (E). Agreement is good for the 20,000 hr^{-1} data (Table 17a), but consistently higher conversion of NO is predicted for 10,000 hr^{-1} space velocity (Table 17b). It is probable that plug flow conditions were not attained at the lower space velocity.

As with the V_2O_5 catalyst, differences in activation energy and the reaction orders between Equation (D) and (E) as well as the small effectiveness factors for the Fe-Cr catalyst indicate the existence of strong pore diffusion effects.

5.2.2. Durability Tests in the Presence of SO_x

A primary objective of this project was to determine the effect of prolonged exposure of the selected catalysts to SO_x. In the previous study it was found that neither catalyst suffered degradation for periods of exposure of about 50 hours. Target periods greater than 100 hours were desired.

The procedure, initially, was to obtain durability data at the standard operational conditions: 20,000 hr^{-1} (STP) space velocity, 400°C reactor temperature, and flue gas containing 1000 ppm NO, 700 to 1100 ppm NH_3, 12% CO_2, 3% O_2, 5% H_2O in N_2 with up to 2500 ppm of SO_x.

5.2.2.1. V_2O_5 Catalyst

The V_2O_5 catalyst was first subjected to a series of intermittent exposure periods of up to 9 hours or less. The intermittent periods were necessitated by the lack of capability, at that time, to handle the large volumes of gases involved. Subsequently, longer exposure periods were employed. Because of the large number of start-up and shut-down periods, each test period with V_2O_5 consisted of initial steady-state operation under SO_x-free conditions. This data provided a reference base for comparison with operation in the presence of SO_x. Steady-state runs with constant SO_x concentrations were then conducted. After each such run, the SO_x flow was terminated and a second steady-state run was conducted in the absence of SO_x. Periodic inlet and outlet concentration measurements were performed during each test period.

Table 18 summarizes the results of the SO_x exposure tests with the V_2O_5 catalyst. The SO_3 present was formed by homogeneous oxidation of the SO_2 in the feed system. The amount of SO_3 formed in the catalytic reactor itself was quite small. Some SO_2 oxidation on the V_2O_5 catalyst, although not alkali-doped as is typical of SO_2 oxidation catalysts, was expected. However, the above results indicate that, under the present conditions, a single, undoped V_2O_5 catalyst bed does not simultaneously reduce NO with NH_3 and oxidize SO_2 with O_2 at 400°C.

The dashes in the beginning of the table represent analyses not performed for the indicated species. In particular, the missing data in several tests include NH_3 analyses of the reactor outlet gas in the presence of SO_2. Even though the SO_2/SO_3 analytical method performed well, some difficulty in obtaining reliable NH_3 data with

TABLE 17a. EXPERIMENTAL AND CALCULATED CONVERSION OF NO ON Fe-Cr (9/1) BASED ON INTRINSIC RATE EXPRESSION (20,000 hr^{-1}).

Temp. (° C)	Inlet Gas Concentrations P^o_{NO} (atm x 10^6)	P^o_{NH} (atm x 10^6)	$P^o_{O_2}$ (atm x 10^6)	Conversion Exptl. (%)	Calculated (%)	η, Effectiveness Factor
200	955	1305	3	12	16	0.45
200	950	1859	3	14	17	0.44
200	740	1340	3	20	18	0.43
200	465	1165	3	17	20	0.37
295	915	723	3	30	37	0.16
295	915	1026	3	31	38	0.16
300	940	957	3	38	38	0.15
300	325	618	3	40	47	0.11
300	480	862	3	38	44	0.12
300	505	536	3	45	43	0.13
300	465	886	4	45	45	0.12
300	460	1084	3	45	43	0.09
300	460	618	3	40	44	0.12
300	745	1002	3	45	41	0.14
300	800	653	3	44	40	0.15
300	760	1328	3	42	41	0.14
300	1375	1375	3	34	36	0.17
300	1375	897	3	33	35	0.17
300	1325	724	3	31	30	0.13
300	1325	2293	3	35	37	0.16
360	555	629	3	59	58	0.07
360	565	758	3	60	59	0.07
360	1030	788	3	62	52	0.09
365	1080	1608	3	56	54	0.08
390	545	1573	3	61	59	0.04
390	1055	1186	3	63	60	0.07
395	965	932	3	69	62	0.07
395	950	1065	3	70	62	0.06
400	1050	1186	3	65	62	0.06
400	1005	690	3	53	61	0.07
400	860	1307	3	70	65	0.06
400	785	1307	3	71	65	0.04
400	890	1017	3	67	64	0.06
400	785	1017	3	66	65	0.06
400	785	762	3	65	64	0.06
400	770	569	3	63	64	0.06
400	1290	1130	3	53	60	0.07
440	1335	1586	3	66	69	0.05

(continued)

TABLE 17a (cont).

Temp. (°C)	Inlet Gas Concentrations P^o_{NO} (atm x 10^6)	$P^o_{NH_3}$ (atm x 10^6)	$P^o_{O_2}$ (atm x 10^6)	Conversion Exptl. (%)	Calculated (%)	η, Effectiveness Factor
440	805	1114	3	70	73	0.03
440	490	884	3	68	74	0.03
440	480	1150	3	71	72	0.03
440	480	1450	3	68	69	0.03
445	985	[illegible]20	3	71	72	0.04
450	945	1162	3	73	74	0.04
445	1020	1198	3	72	72	0.04
450	1140	1198	3	74	72	0.05
485	1335	1586	3	71	77	0.04
490	900	1215	3	74	80	0.02
300	735	545	0.5	32	35	0.17
300	745	506	1	36	37	0.16
300	760	506	3	39	39	0.15
300	750	506	5	42	41	0.14
305	940	799	0.5	28	35	0.17
305	960	799	1	33	37	0.16
305	975	764	3	39	39	0.15
305	1005	787	5	41	40	0.14
305	735	852	0.5	35	38	0.16
305	740	925	1	37	39	0.15
305	840	787	5	43	42	0.14
305	1011	1259	0.5	29	35	0.17
305	1015	1283	1	33	37	0.16
305	1030	1283	3	37	40	0.15
305	1060	1283	5	40	40	0.14
300	1015	1100	1	31	36	0.17
300	1055	1100	5	39	39	0.15
300	770	1053	5	39	42	0.13
395	710	726	1	59	61	0.07
395	760	726	5	70	65	0.06
395	920	1234	0.5	61	58	0.07
395	915	1122	1	62	59	0.07
395	970	1162	5	68	64	0.06
395	950	750	0.5	50	56	0.07
395	960	750	1	53	57	0.08
400	720	908	0.5	58	61	0.07
400	760	905	1	61	62	0.06
400	760	905	3	65	65	0.06
400	800	905	5	67	66	0.06
450	795	905	5	71	76	0.04

(continued)

TABLE 17a (Cont.)

	Inlet Gas Concentrations			Conversion		
Temp. (°C)	P^{o}_{NO} (atm x 10^6)	$P^{o}_{NH_3}$ (atm x 10^6)	$P^{o}_{O_2}$ (atm x 10^6)	Exptl. (%)	Calculated (%)	η, Effectiveness Factor
450	765	905	1	66	72	0.04
445	790	755	1	63	71	0.05
445	790	639	5	70	75	0.04
450	1060	639	5	67	67	0.03
450	1095	905	5	68	73	0.04
450	1105	726	5	63	71	0.05
450	990	1331	1	66	70	0.05
450	1050	1331	5	74	75	0.04
495	1020	1331	1	68	72	0.03
495	1040	1330	5	74	69	0.02
485	780	1235	5	74	80	0.02

Simulated flue gas contains also 12% CO_2, 5% H_2O in N_2.

TABLE 17b. EXPERIMENTAL AND CALCULATED CONVERSION OF NO ON Fe-Cr (9/1) BASED ON INTRINSIC RATE EXPRESSION (10,000 hr^{-1})

	Inlet Gas Concentrations			Conversion		
Temp (°C)	P^o_{NO} (atm x 10^6)	$P^o_{NH_3}$ (atm x 10^6)	$P^o_{O_2}$ (atm x 10^6)	Exptl. (%)	Calculated (%)	η, Effectiveness Factor
250	1080	1005	3	33	41	0.31
250	1065	1005	5	34	44	0.29
250	1120	1105	1	32	37	0.37
350	940	944	1	69	68	0.12
350	900	940	3	69	74	0.10
350	840	944	5	71	74	0.09
400	940	944	1	76	80	0.08
400	950	944	3	78	85	0.06
400	930	944	5	78	87	0.04
310	1055	847	3	58	60	0.16
310	1060	1095	3	58	61	0.16
350	1060	1065	3	72	73	0.11
350	1090	753	3	66	71	0.12
400	1075	1116	3	78	84	0.06

Simulated flue gas contains also 12% CO_2, 5% H_2O in N_2.

TABLE 18. DURABILITY TEST RESULTS - V_2O_5 CATALYST IN THE PRESENCE OF SO_x AT 400° C

Accumulated Time (hr)	Inlet Gas Concentration				Outlet Gas Concentration				Conv. of NO
	NO_x (ppm)	SO_2 (ppm)	SO_3 (ppm)	NH_3 (ppm)	NO_x (ppm)	SO_2 (ppm)	SO_3 (ppm)	NH_3 (ppm)	(%)
Reference	1050	0	0	1160	145	0	0	372	86
2.5	1140	703	0	1160	180	691	--	---	84
7.5	1180	703	0	1160	105	---	--	---	91
Reference	1150	0	0	1089	160	0	0	144	86
9.5	1010	1035	0	1089	115	---	--	86	88
14.5	1060	1035	0	1089	130	---	--	---	88
Reference	1140	0	0	1065	160	0	0	253	86
Reference	1010	0	0	1041	110	0	0	266	89
21.5	995	500	0	1041	115	---	--	---	89
Reference	1090	0	0	1065	105	0	0	291	90
24.5	1090	1489	0	1065	105	1425	113	---	90
30.5	1090	---	--	1065	100	---	---	---	90
Reference	1060	0	0	1114	110	0	0	242	90
39.5	1060	1982	90	1114	175	---	--	---	84
Reference	1150	0	0	1065	130	0	0	411	87
46.5	1140	1000	71	1065	90	---	--	---	92
Reference	1030	0	0	1100	145	0	0	0	86
53.5	1065	917	0	1100	140	---	--	0	87
Reference	1070	0	0	1100	140	0	0	290	87
70.5	1115	1000	--	1100	230	---	--	---	87
Reference	1070	0	0	1020	115	0	0	109	89
99.5	1105	1450	0	914	140	1320	42	470	87
Reference	1030	0	0	1065	115	0	0	---	89
Reference	1050	0	0	1162	145	0	0	372	86
107.0	1180	703	0	1162	105	691	--	---	91
Reference	1150	0	0	1089	160	0	0	144	86

TABLE 18 (cont.)

Accumulated Time (hr)	Inlet Gas Concentration				Outlet Gas Concentration				Conv. of NO_x (%)
	NO_x (ppm)	SO_2 (ppm)	SO_3 (ppm)	NH_3 (ppm)	NO_x (ppm)	SO_2 (ppm)	SO_3 (ppm)	NH_3 (ppm)	
112.0	1060	1035	0	1090	130	-	-	86	88
Reference	1140	0	0	1065	160	0	0	253	86
Reference	940	0	0	1286	165	0	0	310	83
118.5	1090	1011	0	1113	175	1010	51	97	82
Reference	975	0	0	1126	175	0	0	303	82
128.5	1150	892	0	1113	230	820	50	153	79
Reference	900	0	0	1204	145	0	0	315	84
137.5	975	1977	42	918	185	2065	31	250	81
145.0	1125	2583	0	1113	150	2414	52	152	84
Reference	1050	0	0	1574	85	0	0	508	92
163.0	1050	906	0	1586	135	815	48	303	88
Reference	970	0	0	786	260	0	0	61	73
172.0	1050	974	61	858	195	1073	52	60	81
Reference	980	0	0	1145	145	0	0	282	88
176.5	1110	1087	0	1109	145	994	28	97	87
204.5	1070	1086	0	747	120	1088	26	30	88
Reference	1070	0	0	747	145	0	0	145	86
313.0	715	875	31	1225	80	890	42	318	89
480.0	1065	583	52	706	130	554	42	93	87
552.0	1070	620	208	716	30	628	52	--	97
599.0	940	595	219	884	50	658	219	319	97
628.0	1180	553	94	1780	70	407	188	671	94
641.0	1020	0	0	980	130	0	0	436	87
648.0	1005	0	0	710	215	0	0	--	79

S.V. = 20,000 hr^{-1}. Simulated flue gas contains also 12% CO_2, 3% O_2, 5% H_2O in N_2.

the Orion specific ion electrode was experienced. The erratic operation, which was traced to a faulty electrometer used to measure the probe output, was corrected.

For the first 54 hours the usual procedure, as mentioned above, was to conduct a single run of 9 hours or less. During the overnight periods, the catalyst was exposed to flowing air at 400°C. Since these "rest" periods might obscure durability data by providing intermittent catalyst rejuvenation, longer test runs were conducted in subsequent stages. Catalyst deterioration was not observed for test periods up to 32 hours in length.

It should be noted that NO_x and NH_3 concentrations were not precisely the same for reference and durability runs. Data for the durability tests were representative of conditions near the end of a test period. Over extended periods, slight shifts in concentrations occurred, particularly with NH_3, the flow rate of which was subject to the influence of the ambient temperature on the vaporization rate of the liquid NH_3. Nevertheless, the data obtained at reference and run conditions indicate that SO_x did not affect NO_x conversion, which was maintained close to 90% for excess NH_3.

The data indicate reasonable agreement within experimental error between inlet and outlet total SO_x concentrations indicating, based only on these results, that solids formation in the system was either negligible or had, in each case, reached an equilibrium level. On the other hand, the outlet ammonia concentration invariably was lower in the presence of SO_x than in its absence. In most cases, the additional NH_3 usage could not be correlated with corresponding decreases in effluent SO_x. The factors influencing the increased usage of NH_3 in the presence of SO_x have not been resolved, but it is clear that the effect on NO_x conversion is negligible.

It was of interest to determine if operation at lower temperatures would lead to significant catalyst poisoning or salt deposition. Several runs were performed at temperatures as low as 250°C in the presence and absence of SO_x. Table 19 shows typical results with the concentration data given representative of those determined near the end of the test periods. As observed at 400°C, NO_x conversion at lower temperatures was not significantly altered by the presence of SO_x. The results at 250°C were surprising in that the anticipated formation of sulfites and sulfates was expected to "poison" the V_2O_5 catalyst as observed with the platinum catalyst in our earlier EPA program with TRW. The present data implies that classical sulfur poisoning of the Pt catalyst rather than mere salt blockage may have actually occurred in the earlier study although the Pt catalyst could be rejuvenated by heating in air, at least after short exposure to SO_x.

At the lower temperatures, significant oxidation of SO_2 to SO_3 on V_2O_5 occurred, but it is not certain if the effect is catalytic or homogeneous. Formation of SO_3 is thermodynamically favored over the entire temperature range employed and for the reaction,

TABLE 19. DURABILITY TEST RESULTS - V_2O_5 CATALYST IN THE PRESENCE OF SO_x AT TEMPERATURES BELOW 400°C.

Accumulated Time (hr)	Temp. (°C)	Inlet Gas Concentration NO_x (ppm)	Inlet Gas Concentration SO_2 (ppm)	Inlet Gas Concentration SO_3 (ppm)	Inlet Gas Concentration NH_3 (ppm)	Outlet Gas Concentration NO_x (ppm)	Outlet Gas Concentration SO_2 (ppm)	Outlet Gas Concentration SO_3 (ppm)	Outlet Gas Concentration NH_3 (ppm)	Conv. of NO_x (%)
204.5		Accumulated to start of series of tests								
Reference	255	880	0	0	862	410	0	0	404	53
220.5	250	955	1188	0	859	440	702	528	153	54
232.0	250	980	1188	0	859	400	---	---	---	59
Reference	300	865	0	0	820	155	0	0	147	82
236.0	300	870	1024	0	845	130	575	539	140	85
Reference	255	1035	0	0	770	470	0	0	323	55
238.0	255	1075	1100	0	858	500	567	507	28	53
243.5	300	1085	1100	0	858	295	---	---	---	73
267.0	350	1000	1100	0	858	140	---	---	---	86
272.0	350	840	1100	0	858	90	---	---	---	89
Reference	355	890	0	0	768	170	0	0	27	81

---

s.v. = 20,000 hr^{-1}. Simulated flue gas contains also 12% CO_2, 3% O_2, 5% H_2O in N_2.

$$SO_2 + \frac{1}{2} O_2 \rightarrow SO_3$$

K_p increases approximately by two orders of magnitude when temperature is decreased from 400 (K_p = 600) to 250°C (K_p = 85,000). The failure to observe significant SO_3 at the reactor inlet when the mixer-vaporizer temperature was reduced to 250°C (data not shown) suggests that below 400°C, the oxidation may be catalyzed on the V_2O_5.

Immediately after introduction of SO_x, NO_x conversion decreased (10% or less). Recovery of initial activity appeared to be a function of temperature; at 250°C recovery took 2 to 5 hours while at 400°C recovery occurred in a matter of minutes. The data in Tables 18 and 19 are steady-state results for each test period. Additional data other than NO_x conversion were not obtained during the recovery period. Tentatively, the observed induction period for the recovery of activity implies that, initially, formation of salts somewhere in the test system may have occurred, depleting the NH_3 supply. Recovery indicates the attainment of a steady-state or equilibrium quantity of the salt in accord with the satisfactory balances between steady-state inlet and outlet SO_x as discussed earlier.

After about 200 hours, the gas handling capabilities of the laboratory were expanded to enable long periods of continuous runs. During the overnight periods, the catalysts were exposed to simulated flue gas containing 500 ppm NO, 500 ppm NH_3 and 350 ppm SO_x at 400°C.

Figure 31 shows that during the SO_x exposure period the conversion of NO_x was consistent with pre-exposure data (the solid line is representative of pre-exposure data). Also shown in the figure are several data points taken in the absence of SO_x after 600 hours of exposure to SO_x. In general, no change in activity was evident. The total exposure period for the durability test of the V_2O_5-Al_2O_3 catalyst was 648 hours.

5.2.2.2. Fe-Cr Catalyst

As with the V_2O_5 catalyst, conversion data of the 9/1 Fe-Cr catalyst durability tests were accumulated with the standard flue gas composition with either stoichiometric or excess NH_3. The Fe-Cr catalyst was also normally exposed overnight to simulated flue gas containing 500 ppm NO, 500 ppm NH_3 and 350 ppm SO_x at 400°C.

Table 20 summarizes the conversion data obtained during the durability tests at 400°C. No significant degradation of catalyst performance was observed during the 1052 hours test.

The stability of the Fe-Cr catalyst is markedly different from the iron oxide-chromium oxide catalyst studied by Matsuda et al (5-2). Their catalyst preparation method was not specified (it may have followed the patent of Nonnenmacher and Kartte (3-1)). In their study deterioration began immediately after exposure to 500 ppm SO_3.

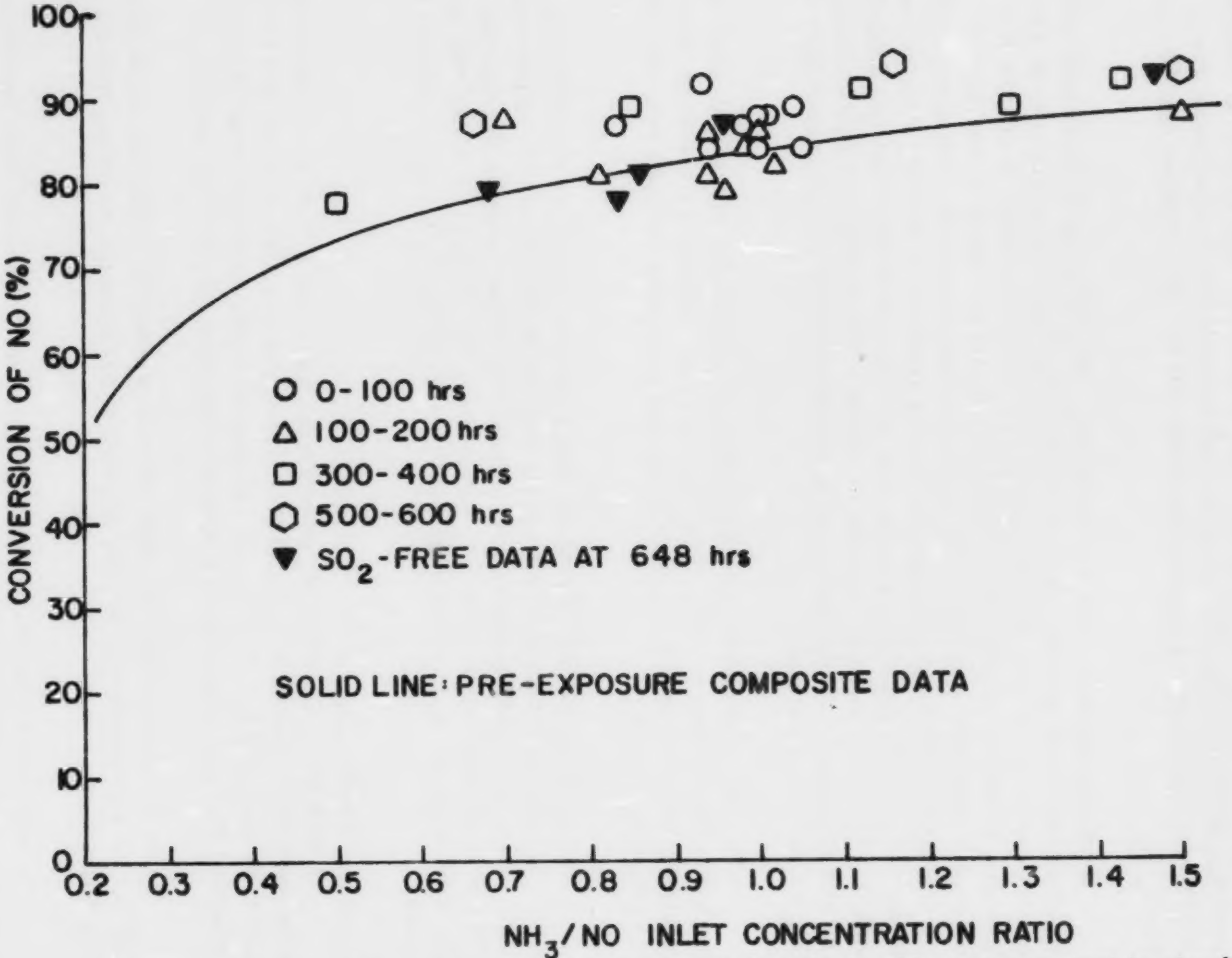

Figure 31. Conversion of NO on V_2O_5 catalyst during SO_x exposure test in simulated flue gas (1000 ppm NO) at 400°C. s.v. = 20,000 hr^{-1}.

TABLE 20. DURABILITY TEST RESULTS - Fe-Cr CATALYST IN THE PRESENCE OF SO_x AT 400°C

Accumulated Time	Inlet Gas Composition				Outlet Gas Composition				Conv. of
	NO_x	SO_2	SO_3	NH_3	NO_x	SO_2	SO_3	NH_3	NO
(hr)	(ppm)	(ppm)	(ppm)	(ppm)	(ppm)	(ppm)	(ppm)	(ppm)	(%)
reference	1160	0	0	670	470	0	0	0	60
0.5	1130	419	225	670	480	521	159	16	61
20	1060	---	---	---	420	---	---	--	60
45	1050	---	---	---	415	---	---	--	60
62	1045	578	173	668	405	549	176	58	63
70	1130	---	---	---	440	---	---	--	61
88	1110	720	187	1184	275	673	259	30	75
94	1110	---	---	---	265	---	---	--	76
136	1095	1044	62	1106	360	764	83	33	67
140	1025	---	---	---	340	---	---	--	67
159	1025	570	52	1368	275	653	20	76	74
182	1115	673	52	1184	260	634	52	91	77
189	985	---	---	---	255	---	---	--	74
206	1070	704	73	1184	230	258	83	19	79
303	1100	---	---	---	280	---	---	--	75
346	1075	554	41	976	315	474	83	43	71
417	1105	673	156	916	295	669	69	34	73
422	1090	---	---	---	300	---	---	--	73
440	1045	669	61	1036	160	600	21	112	85
448	1060	---	---	---	160	---	---	--	85
reference	995	0	0	808	285	---	---	--	71
472	1010	665	61	976	245	609	104	38	76
478	1095	---	---	---	270	---	---	--	75
reference	1000	0	0	903	310	0	0	0	69
480	1030	574	61	916	330	504	105	27	69
485	1040	---	---	---	320	---	---	--	69
504	1000	377	167	667	385	340	192	31	62
510	1050	---	---	---	320	---	---	--	70
526	1085	518	52	736	350	382	188	48	68
533	1000	---	---	---	350	---	---	--	65
597	960	449	133	1036	215	380	177	127	78
613	1000	390	167	916	155	497	31	55	84
620	1010	---	---	---	175	---	---	--	83
637	995	589	104	1183	150	653	31	71	86
715	1050	455	37	652	390	409	57	390	63
718	1060	---	---	---	380	---	---	--	64
732	985	953	83	634	360	1023	65	66	64

(continued)

TABLE 20 (cont.)

Accumulated Time (hr)	Inlet Gas Composition NO_x (ppm)	SO_2 (ppm)	SO_3 (ppm)	NH_3 (ppm)	Outlet Gas Composition NO_x (ppm)	SO_2 (ppm)	SO_3 (ppm)	NH_3 (ppm)	Conv. of NO (%)
736	980	---	---	634	360	----	---	--	63
754	1000	945	187	706	305	1031	135	54	70
763	1020	---	---	---	310	----	---	--	70
776	1000	1430	60	618	340	1497	25	36	66
784	1045	----	---	---	320	----	---	--	69
801	900	1113	52	757	275	1051	63	48	70
811	940	----	---	---	280	----	---	--	70
reference	900	0	0	710	315	0	0	37	65
873	930	1251	94	639	315	1177	52	51	66
897	935	1153	63	682	270	1158	37	47	71
923	940	1157	59	757	250	1164	26	36	74
945	920	967	68	635	255	1028	59	41	72
997	1000	920	89	706	235	966	94	43	77
1002	900	1067	19	690	205	970	64	33	77
reference	910	0	0	1077	125	0	0	98	86
1018	885	929	41	1032	135	880	52	51	85
*1045	975	1185	84	758	280	1091	48	--	71
1050	1115	1170	511	660	—	566	522	190	--
1052	1280	0	0	1373	280	0	0	400	78

20,000 hr^{-1} space velocity. Simulated flue gas contains also 12% CO_2, 3% O_2, 5% H_2O in N_2.

* 350 C

The catalyst recovered activity at 300 hours and then again deteriorated rapidly. At about 800 hours the catalyst was about 65% as effective as before exposure. Matsuda et al (5-2) attributed the deterioration of activity to the formation of iron sulfate from the reaction of the catalyst with SO_3.

In the present study, the inlet gas containing sulfur oxides was predominately SO_2 with SO_3 concentrations roughly about 100 ppm on the average. Since the SO_3 concentrations in the Matsuda study were higher (500 ppm), a direct comparison of degradation effects may not be valid. However, there is no evidence for the direct reaction of SO_3 and Fe_2O_3 on the UCLA Fe-Cr catalyst since inlet and outlet SO_3 concentrations were approximately equal and total SO_x balances were, in most respects, reasonably close as shown in Table 20. Furthermore, several catalysts disclosed in the patent literature (see Appendix A) for the removal of NO with NH_3 involve pre-sulfation of the catalysts with either aqueous sulfates or oxides of sulfur. Therefore, there is some evidence that certain metallic sulfates are active catalysts.

The degradation of the Fe-Cr catalyst in the Matsuda study may be due to SO_3-carrier reaction. However, since details of the catalyst formulations in their work were not provided, an external evaluation of their degradation data is precluded. Nevertheless, SO_3-alumina reaction is a possible cause for degradation of alumina-supported catalysts. The extent and induction time for degradation should depend on the type of active material and the ratio of active material to alumina. Such information does not appear to be available, at least in the open literature.

Figure 32 shows that there was no overall decrease in the conversion of NO. There was, in fact, the indication that the catalyst activity may have increased somewhat (the solid line is a composite of data for pre-SO_x exposure tests), particularly at low NH_3/NO ratios (<0.65).

Typical data obtained during the overnight test periods with, nominally 500 ppm NO, 400 ppm NH_3 and 350 ppm SO_x at 400°C are given in Table 21. Space velocity was maintained at 20,000 hr^{-1} (STP). Typical conversions ranged from 85-92%. These percent conversions were considerably above the 400°C curve of Figure 26 for comparable conditions studied before the catalyst was exposed to SO_x.

Although Figure 32 shows, for higher NO and NH_3 concentrations, a modest increase in catalytic activity after initial exposure to SO_x, Table 21 indicates that, at lower NO and NH_3 concentrations, conversion of NO is markedly increased. The third-to-last entry of the table, (SO_x-free conditions) indicates that the improved conversion over that observed prior to SO_x exposure does not appear to be related to the immediate presence of SO_x itself but is a cumulative change in the catalyst.

The improvement in activity with aging in the presence of SO_x for both 1000 and 500 ppm NO indicates that the rate expressions, which were based on the parametric studies, apply to relatively fresh catalysts. For well-aged catalysts, the conversion rates calculated from the developed rate expressions would give lower bound values.

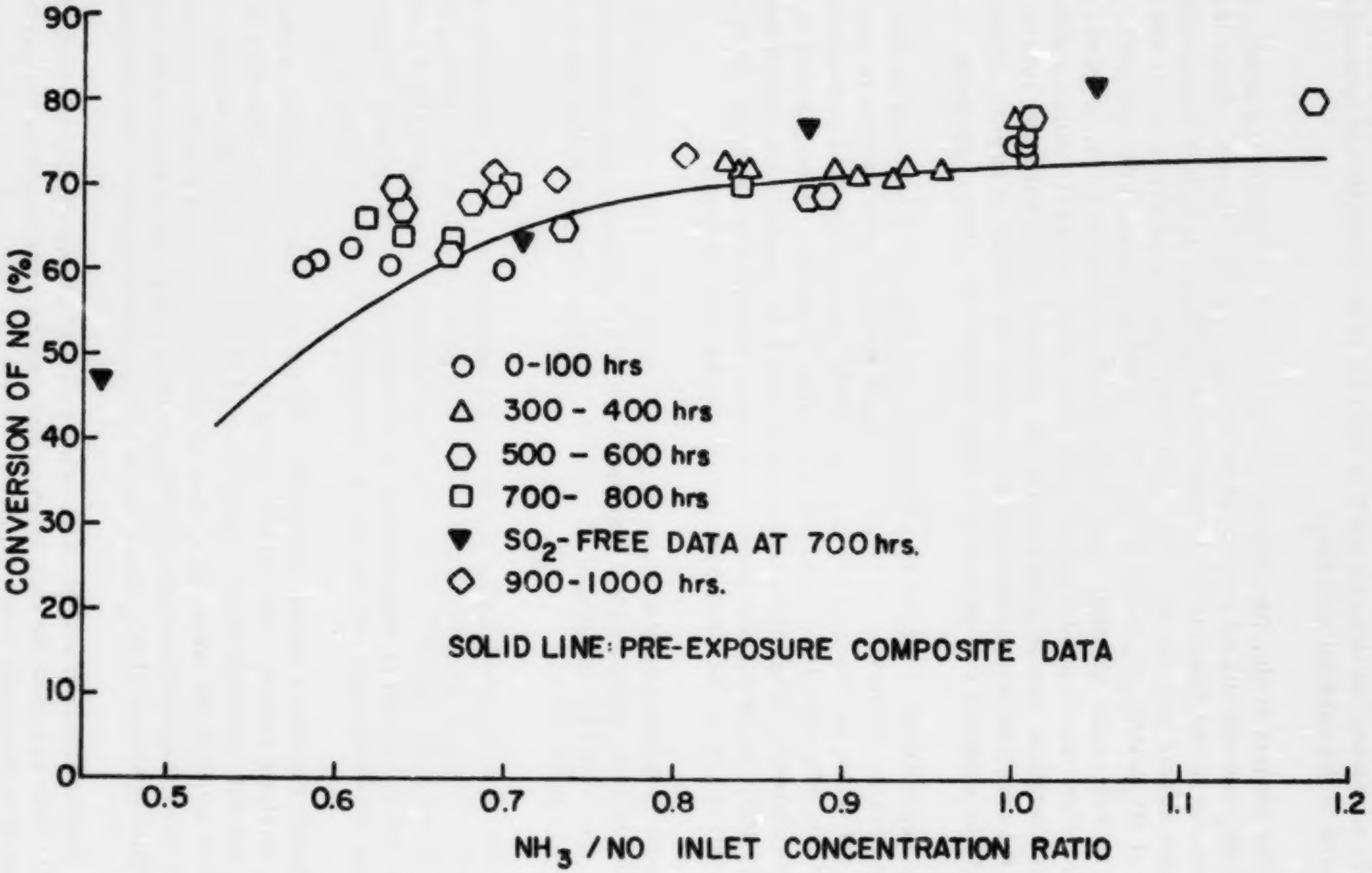

Figure 32. Conversion of NO over Fe-Cr catalyst during SO_x exposure test period in simulated flue gas (1000 ppm NO), 400^{o}C, (s.v.=20,000 hr^{-1}).

TABLE 21. CONVERSION OF LOW CONCENTRATIONS OF NO IN PRESENCE OF SO_x AT 400°C OVER Fe-Cr CATALYST

Accumulated	Inlet Gas Composition				Outlet Gas Composition				Conv. of
Time	NO_x	SO_2*	SO_3	NH_3	NO_x	SO_2	SO_3*	NH_3	NO
(hr)	(ppm)	(ppm)	(ppm)	(ppm)	(ppm)	(ppm)	(ppm)	(ppm)	(%)
730	540	305	--	323	65	---	--	0	88
753	540	331	--	456	100	270	--	0	82
775	510	269	42	482	80	262	37	-	84
800	510	281	--	500	105	---	--	-	80
892	310	466	--	639	25	---	--	-	92
893	450	369	84	481	65	388	52	-	86
918	535	425	31	336	75	---	--	-	86
943	470	238	44	468	75	---	--	-	84
965	450	0	0	465	70	0	0	-	84
968	490	481	83	310	75	---	--	-	85
1014	460	237	68	659	75	241	52	-	87

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* Where dashes appear in SO_3 column, the SO_2 data include SO_3 formed in in the feed system (i.e., for these cases SO_2 actually represents SO_x).

20,000 hr^{-1} space velocity. Simulated flue gas contains also 12% CO_2, 3% O_2, 5% H_2O in N_2.

Investigation of interaction of SO_3 and Al_2O_3 or other carrier material is important. The concentrations of SO_3 present in this project may have been too low to induce catalyst degradation effects. Additional effort in this area either concurrently or before advancement to demonstration scale of operation is highly desirable. Identification of the compound(s) formed and crystallinity changes during prolonged exposure to higher concentrations of SO_3, and methods to inhibit the degradation effects as well as identification of carrier materials immune to the presence of SO_3 are particularly important problems to be resolved.

5.2.3. Recommended Operating Conditions

Removal of large concentrations of NO (1000 ppm or greater) with NH_3 in excess can be achieved on both catalysts at the 80% level or higher at a space velocity of 20,000 hr^{-1}. Removal of approximately 90% for lower NO concentrations has been observed at that space velocity. At 10,000 hr^{-1}, 90 to 100% removal of NO is obtained.

An estimate of the capital and operating costs of non-noble metal catalysts as adapted to an 800 MW power plant operating at 400°C and 15,000 hr^{-1} space velocity has been made previously (1-2). Working capital and investment return have been excluded. By assuming 10-year depreciation, and annual catalyst replacement, the estimated costs (1973 dollars) are:

capital cost — \$6.15/KW

operating cost — 0.088 ¢ / KWH

The most significant operating expense was, as expected, the cost of the NH_3 feed. Since catalyst rejuvenation/replacement was estimated to be less than 5% of both the operating and the capital cost (initial charge), differences in the costs of the V_2O_5 and Fe-Cr catalysts do not change significantly the total capital and operating costs

5.3. Task 3. INVESTIGATION OF NEW AND PROMISING CATALYSTS

The primary source of literature on NO_x catalysis was a computerized search provided by the UCLA library which was updated at biweekly intervals. The latter was supplemented by compilations obtained from APTIC. Particular emphasis was placed on selective reduction of NO_x with NH_3, although the references were scanned for claims of selectivity in the presence of O_2 with other reducing agents.

Applicable literature listings, as well as pertinent data (when available) such as catalyst type, test conditions, and poisoning effects are tabulated in Appendix A along with an overall bibliography (from which the catalyst list was assembled). Acquisition of all the pertinent complete articles and patents was not possible during the course of this project. Most of the tabulated data were obtained from published abstracts rather than the complete articles.

The primary source of the literature was from patent filings and many of these were Japanese disclosures. Several of the patents were obtained and translated. Preliminary inspection of the patent literature reveals that a large number of patents related to the basic disclosures of Nonnenmacher and Karrte (3-1) for vanadia, molybdena and tungsten oxide catalysts and of Schmidt and Schulze (3-2) for iron-chromium catalysts. It is beyond the scope of the present project to explore all the variations in preparation of these catalyst types.

Several catalysts were of considerable interest and samples were prepared for brief comparative studies. Two of the catalysts were reported by Hitachi Ltd. (5-3) to be active for NO_x reduction with NH_3 at 250°C in the absence of SO_2 and at about 350°C in the presence of SO_2. Both catalysts (5-4 and 5-5) apparently contain mixed oxides of the general formula $La_2Cu_{1-x}Zr_xO_4$, which has a crystal structure similar to K_2NiF_4. The two catalysts were prepared in our laboratory by dry pressing of the mixed oxide with Al_2O_3 (5-4) and by impregnation methods (5-5), respectively.

A third catalyst of interest was prepared by substitution of a portion of the V_2O_5 catalyst with an inexpensive active material in an attempt to reduce catalyst cost without a loss in performance. The demonstrated high activity of iron-containing catalysts suggested a binary mixture of V_2O_5 and Fe_2O_3. Consequently, a catalyst was prepared by impregnation of Al_2O_3 (Filtrol 86) with a solution of $Fe(NO_3)_3 \cdot 9\,H_2O$ and NH_4VO_3 with oxalic acid added to increase the solubility of the latter (probably by reduction of V^{+5} to V^{+4}). The impregnated material was then calcined at 500°C. Final total oxide concentration (Fe: V ratio of 1/1) was 15% .

A fourth catalyst was prepared by impregnating Al_2O_3 (Filtrol Grade 86) with a solution containing NH_4VO_3 and $H_2C_2O_4$ (2:1 molar ratio), $Fe(NO_3) \cdot 9\,H_2O$ and CrO_3 to obtain a 10% Fe-Cr-V (18/1/1) catalyst. After impregnation, the catalyst was calcined in air for 16 hours at 500°C.

The two La-Cu-Zr catalysts (Hitachi #1, ref. 5-4 and Hitachi #2, ref 5-5), the Fe-V and the Fe-Cr-V catalysts, were tested from 200 to 500°C with an NH_3/NO ratio of 1.2 in simulated flue gas containing 1000 ppm NO. Figure 33 shows the results of the tests and includes comparative data for the optimum 15% V_2O_5 and 10% Fe-Cr (9/1) catalysts. The Fe-V catalyst was tested only at 20,000 hr^{-1} space velocity while the catalysts prepared from the Hitachi patents and the Fe-Cr-V catalyst were tested at 10,000 hr^{-1} space velocity.

Figure 33 shows that the Fe-Cr-V catalyst was the most active followed by the Fe-V, V_2O_5 and then the Fe-Cr catalysts. The maximum conversion occurred at 400°C for the four UCLA catalysts. On the other hand, for the Hitachi catalysts, the maximum conversion occurred at the lower temperature of 330°C, but the magnitude of NO_x conversion was substantially lower than the other four catalysts.

In view of its superior activity, a durability test of about 180 hours in the presence of SO_x was performed on the Fe-Cr-V catalyst. Table 22 shows that the presence

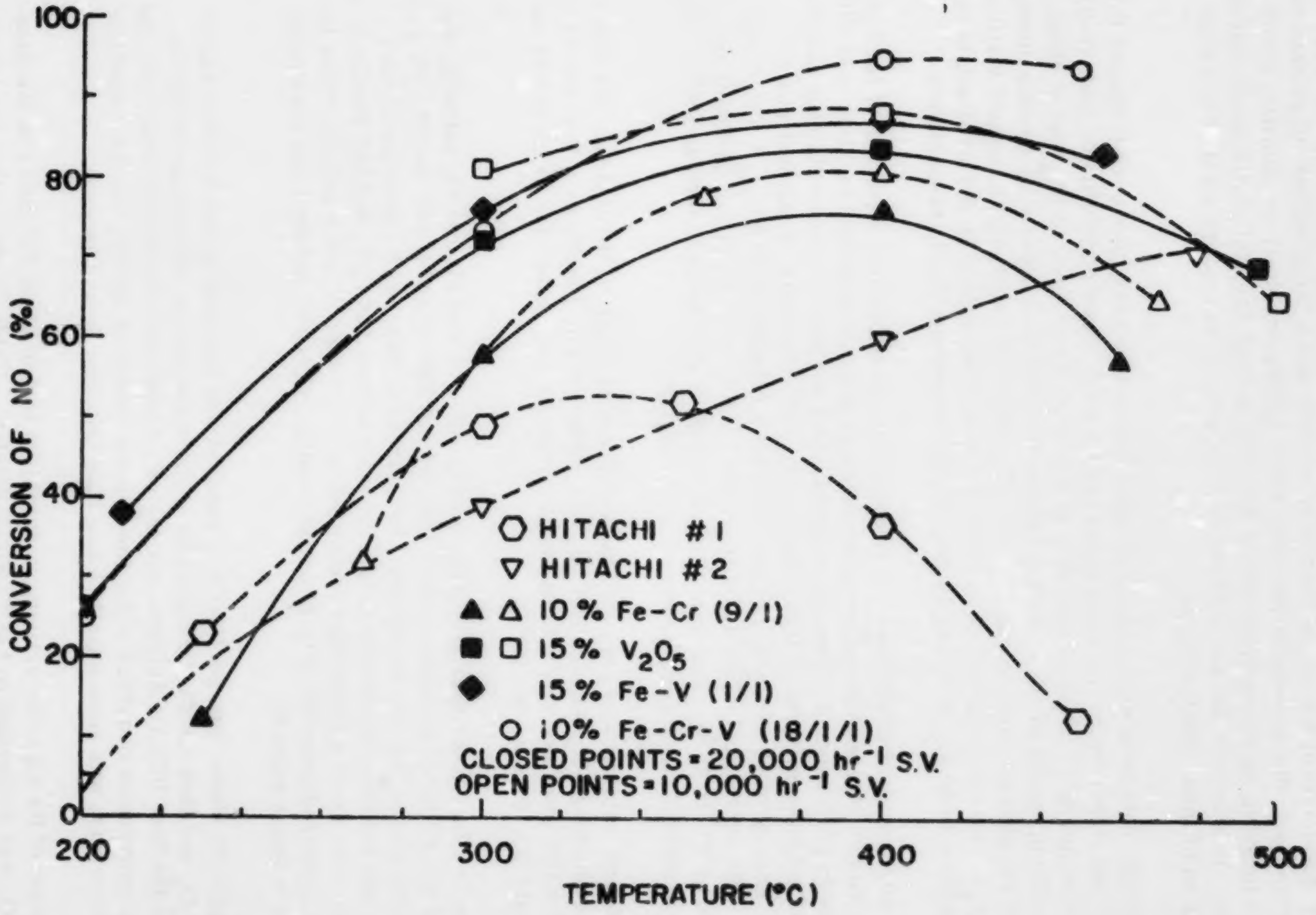

Figure 33. Comparison of several active catalysts for NO reduction with NH_3 in simulated flue gas. 1000 ppm NO, 1200 ppm NH_3.

of SO_x increased activity for removal of NO with NH_3 in simulated flue gas. For the duration of the test (186 hours), degradation of the catalytic activity was not observed.

TABLE 22. DURABILITY TEST RESULTS - 10% Fe-Cr-V (18/1/1) CATALYST IN THE PRESENCE OF SO_x AT 400°C.

Accumulated Time (hr)	Inlet Gas Composition				Outlet Gas Composition				Conv. of NO (%)
	NO_x (ppm)	SO_2 (ppm)	SO_3 (ppm)	NH_3 (ppm)	NO_x (ppm)	SO_2 (ppm)	SO_3 (ppm)	NH_3 (ppm)	
reference	1055	0	0	1257	50	0	0	--	95
56	950	275	110	1450	10	320	96	671	99
80	990	828	78	952	60	755	92	591	94
100	580	280	104	494	10	320	140	206	98
128	1240	420	590	884	--	--	--	--	--
133	1040	905	30	1347	20	--	--	--	98
163*	740	35	43	957	190	35	43	629	74
186	1120	233	136	822	20	222	153	--	98

s.v. = 10,000 hr^{-1}. Simulated flue gas contains also 12% CO_2, 3% O_2, 5% H_2O in N_2.
* Catalyst taken out of reactor and exposed to air for two days after 142 hours exposure time.

A very brief durability test of the 15% Fe-V (1/1) - Al_2O_3 catalyst in simulated flue gas (s.v. = 10,000 hr^{-1}) containing SO_x (420 ppm SO_2, 110 ppm SO_3) was conducted at 400°C. Concentrations of NO_x and NH_3 were 1040 and 1210 ppm, respectively. At the end of the 26 hour exposure test, 99% conversion of NO_x was obtained.

The promising results for the Fe-Cr-V and the Fe-V catalysts indicate that further tests are warranted including modification of catalyst composition and catalyst loading to improve activity and to reduce costs. In addition, parametric studies and long term durability tests in the presence of SO_x (including concentrations of SO_3 > 500 ppm) are recommended.

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APPENDIX A

Specific references of direct pertinence to this project have been examined and are listed in Table A-1 which includes, in addition to the normal bibliographic notations, the institution or company affiliations and the specific Chemical Abstracts reference number. Data, which are pertinent to NO_x catalysis, obtained from these abstracts and/or papers have been assembled and presented in Table A-2. Specific information presented is catalyst type and composition, test conditions, conversion of NO_x or outlet concentrations, selectivity in the presence of O_2, resistance to sulfur oxides and the extent of durability tests, if reported. Data for reducing agents other than NH_3 (these are predominantly concerned with automotive NO_x abatement systems or, in a few cases, with HNO_3-plant tail-gas treatment) are also listed in Table A-2. The listings suggest that much of the activity in NO_x catalysis is by Japanese workers and indicate, surprisingly, a lesser interest in the field by U.S. industry and governmental agencies. The bibliographical listing covers the approximate period from June 1974 to the present and can be considered to be an update of the previous survey (1-2).

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TABLE A-2. PERTINENT DATA IN REFERENCES ON NO_x CATALYSIS

Catalyst	Company/Institution	Reductant	Operating Conditions
CuO 95-10, NiO 5-90 on Al_2O_3	Mitsubishi Metal Mining Co.	CO/butane	1200 ppm NO, 350 ppm C_4H_{10}, 1.9% CO, 0.65% O_2, 5.8% CO_2, 8% N_2 on N_2. 12000 hr^{-1} S.V., 500-520°C.
> 0.1 Ta or Nb on ZrO_2	Kaneguchi Chemical	None	20000 ppm NO. 800°C.
$Sr_x (R.E.)_{1-x} NiO_3$ on SiO_2 R.E. = Rare Earth $x = 0.01 - 0.9$	Hitachi Makuseru	N.R.	N.R.
WO_2	Mitsubishi Chemical	None	> 500°C
4th Period Metal Oxides	Inst. Katal., Novosibirsk, USSR	CO	Above 140°C.
Ni 40-95, Cu 5-60 on Cr - Stainless Steel	Esso Research	CO	Auto Exhaust
Cu, Cr, Mn, Fe, Co Oxides on C	Toray Industries	None	0.47% NO in N_2
K, Na, Cu, Zn (1-20%) on Fe_2O_3	Kharkov Polytechnic USSR	N.R.	N.R.
Ru on ceramic	Gulf Research	CO, H_2, HC	204 - 816 °C Auto Exhaust
Ba. Nd(Ba/Nd = 1 : 10 - 1 : 100) on ZrO_2	Ethyl Corp.	None	> 400°C
Ba - Cu Chromate	Daimler Benz	CO, HC, H_2	Auto Exhaust
$R.E._{2-x} B_x NiO_4$ on various supports	Hitachi Ltd.	CO	1000 ppm NO, 4000 ppm CO 500°C
Cu, Ni, Cr, Mn, Mo on Carbon	Nissan Engr.		60,000 ppm NO, low temperature in a column containing water or HNO_3.

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
94%	NR	NR	NR	1	
65 ppm	NR	NR	NR	3	
NR	NR	NR	NR	5	
NR	NR	NR	NR	6	
---	NR	NR	NR	7	Activity order: $Co_3O_4 > SnO_2 \sim Cr_2O_3 > Fe_2O_3 > NiO \sim Mn_2O_3 > ZnO >> TiO_2$ O_2O_5, MgO.
NR	NR	NR	NR	9	
< 90%	NR	NR	NR	10	Probably the reaction involved the carbon carrier.
NR	yes	NR	NR	11	
NR	NR	NR	NR	12	
NR	NR	NR	NR	14	
NR	NR	NR	NR	15	
100%	No	NR	NR	16	
1200 ppm	NR	NR	NR	17	Concentration of HNO_3 3-phase, heterogeneous system.

(cont.)

TABLE A-2 (cont.)

Catalyst	Company/Institution	Reductant	Operating Conditions
Ce/Ni on ZrO_2	Hitachi Maxell	CO	0.5% NO, 2% CO in "waste" gas, 300°C at 20,000 hr^{-1}
Na_2O or K_2O or Cs_2O on Al_2O_3	Mitsubishi Chemical	CO, HC, H_2	HNO_3 plant tail gas or auto exhaust 700°C.
SnO_2 - CuO (Cu : Sn = 0.5-0.6 : 1)	Tin Res. Inst. England	CO	150°C
50% Cu - exchanged mordenite + 50% Ni-exchanged mordenite		CO, HC, H_2	400 - 480°C in auto exhaust
Steel with Cu + Al_2O_3 outer layer	Toyota Motor Co.		Auto Exhaust
Copper-Nickel Alloy	Ube Industries	CO, H_2	2000 ppm NO, 12% CO, 5% H_2, 1% O_2, 7% H_2O, 68% N_2 at 100,000 hr^{-1}
Copper - Al_2O_3 (8% CuO)	Esso Research and Engr.	NH_3	1000 ppm NO_x, 2700 ppm SO_2 3-5% O_2, 5000 hr^{-1} $NH_3/NO = 0.7 - 0.8$.
Copper - Al_2O_3 (1.45% Cu)	Esso Research and Engr.	NH_3	553 ppm NO_x, 0.25 - 0.3% SO_2, 6.8% O_2, fly ash, 2000 hr^{-1}, $NH_3/NO = 3.3$
Ag x Zeolite	USAEC		1% NO_2, 1.9% NH_3 in air 0.15 sec residence time
$La_{2-x}A_xCu_{1-y}-B_yO_4$ (See remarks) on ceramic oxides	Hitachi, Ltd.	CO, H_2	"combustion" exhausts
Rh (.001 - 5%) on refractory	Imperial Chemical	CO, H_2	Exhaust gas with A/F ratio of 0.95-1, 20,000-200,000 hr^{-1}

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
500 ppm	NR	NR	NR	18	
76%	NR	NR	NR	19	
High	NR	NR	NR	21	
High	NR	NR	NR	25	
NR	NR	NR	NR	26	
(300°C) 84 % (350) 99 % (400) 100 % (450) 100 %	No	NR	NR	27	
(399°C) 100 %	Yes	Yes	NR	28	
(360°C) 15 ppm	Yes	Yes	NR	29	Catalyst absorbs SO_2 and is regenerated by reduction in H_2
(200-250°C) 99 %	Yes	NR	NR	30	
(700-800°C) High	NR	NR	NR	33	A = Y, alkaline earth or lanthanide B = Li, Al, Ti, V, Cr, Mn, Fe, Co, Ni, Zn, Ga, Zr, Sn, Mo or W $0 \leq x \leq z$, $0 \leq y \leq 1$
NR	NR	NR	NR	34	

(cont.)

TABLE A-2 (cont.)

Catalyst	Company/Institution	Reductant	Operating Conditions
$Re_{1-x}M_xM'O_3$ (see remarks) on Al_2O_3	Western Electric	H_2	Auto Exhaust
$A_xB_{1-x}MO_3$ (see remarks) on ZrO_2	Hitachi Maxell	NR	Waste Gas Cleaning
CeO_2 - Co_2O_4 on ZrO_2	Hitachi Maxell	NR	NR
$A_{1-x}B_xCo_{1-y}$ - Ni_yO_{5-d} (see remarks)	Hitachi, Ltd.	NR	NR
$SrLaNiO_4$	Hitachi Maxell	NR	NR
CuO (0.1 - 10% as Cu) La_2O_3 (0.1 - 20% as La) ZrO_2 (0.01 - 20 as Zr) on ceramic carrier	Hitachi, Ltd.	CO, H_2 (also can add NH_3)	Exhaust Gas
Pd, Pt or Rh doped Ni or CeO_2 on ZrO_2	Hitachi Maxell	CO	0.5% NO_x, 2% CO, 2000 hr^{-1}
Ag x Zeolite	Pence & Thomas	NH_3	1% NO_2, 1.9% NH_3 in air
Cu, Pt, Pd, Ru or other transition elements on oxides of alkaline earths or alkalis or Al_2O_3	Hitachi Shipbuilding	CO	400-500 ppm NO_x, 1.1% CO, 0.3% O_2, 1.3% CO_2, 9% H_2O, 10,000 hr^{-1}
Ni and CeO_2 on NiO + ZrO_2 carrier	Hitachi Maxell	CO	0.5% NO_x, 2% CO, 3% H_2O, 20,000 hr^{-1}

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
(100% at T > 300°C) NR	NR	NR	NR	35	Re = Rare Earth, M = Ba, Ca, Pb and/or Sr, M' = transition metal, $x = 0.05 - 0.7$
97% NR	NR	NR	NR	39	A = Alk. Earth, B = Rare Earth, M = Co, Mn or Ni, $x = 0.01 - 0.9$
NR	NR	NR	NR	40	
NR	NR	NR	NR	41	$0.1 \leq x \leq 1.0$, $0.015 \leq y \leq 1.0$, $0 \leq d \leq x/2$, A = Lu or Nd, B = Sr or Ca
NR	NR	NR	NR	42	
99%	NR	NR	NR	43	
(300°C) 2000 ppm NO_x	NR	Yes	NR	44	
(200-250°C), <0.1 ppm NO_2, <0.1ppm NO	Yes	NR	NR	45	
(400°C) 100%	NR	NR	NR	48	
(400°C) 400 ppm NO_x	NR	NR	NR	50	

(cont.)

TABLE A-2 (cont.)

Catalyst	Company/Institution	Reductant	Operating Conditions
Ni, CeO_2 and Li_2O on ZrO_2	Hitachi Maxell	CO	0.5% NO_x, 2% CO, 20,000 hr^{-1}
CeO_2 or ClO_3 (10%) on Al_2O_3	Mitsubishi Chemical	NH_3	2000 ppm NO_x, 3000 ppm NH_3 10% O_2, 40,000 hr^{-1}
Pd and Rh - doped NiO on cordierite	W.R. Grace	CO	1000 ppm NO, 250 ppm C_3H_6, 2% CO, 0.2 - 1% O_2, 10% CO_2, 10% H_2O 100,000 hr^{-1}
Polyacrylonitrile on SiO_2	British Leyland	CO	1000 ppm NO, 250 ppm C_3H_6, 2% CO, 0.2 - 1% O_2, 10% CO_2, 10% H_2O 100,000 hr^{-1}
Cu wool containing Al_2O_3 and C	Fujikura Cable Works	CO	900 ppm NO, 4.5% CO, 25% O_2
$CuSO_4$ on porous oxide	Hitachi, Ltd.	NH_3	730 ppm NO, 950 ppm SO_2, 490 ppm NH_3, 1% O_2, 12% CO_2, 12% H_2O 23,000 hr^{-1}
Cu or Ni oxides on Cu alloy	Fujikura Cable Works	CO	700 ppm NO, 2% CO, 0.5% O_2
Ni - CeO_2 - Bi on ZrO_2	Hitachi Maxell	CO, H_2	1500 ppm NO, 700 ppm HC, 1.5% CO, 1.5% H_2, 0.5% O_2, 12% CO_2, 12% H_2O, Temp. not reported
Fe_3O_4 (5%) and MnO_2 (95%)	Toray Industries	Oxidation	0.43% NO, 2.01% O_2, 1.6% H_2O
CuO	Esso Research and Engr.	NH_3	553 ppm NO, 0.25 - 0.30% SO_2, trace SO_3 in stack gas, 2000 hr^{-1} NH_3/NO=3.3-3.6; 360°C.
CuO (2.8%) on Al_2O_3	Hitachi, Ltd.	NH_3	735 ppm NO, 800 ppm SO_2, 570 ppm NH_3, 1% O_2, 12% H_2O, 12% CO_2, 350°C.

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
(300°C) 300 ppm NO_x	NR	NR	NR	51	
(360°C) 94%	yes	NR	NR	62	
(593°C) 95%	no	NR	NR	63	
(600°C) 90-98%	no	NR	NR	64	
91%	see remarks	NR	NR	65	Apparent selectivity may be due to reaction of O_2 with C.
(300-500°C) 100%	yes	yes	NR	66	
(680°C) 90%	no	NR	NR	67	
150 ppm NO	no	NR	NR	68	Produced 450 ppm NH_3
Performance not reported	--	NR	NR	69	
15 ppm NO	yes	yes	NR	70	Process description involving multiple beds of decreasing CuO content absorbed on catalyst.
100%	yes	yes	NR	72	

(cont.)

TABLE A-2 (cont.)

Catalyst	Company/Institution	Reductant	Operating Conditions
MnO_2 - Co_3O_4 (95:5)	Toray Industries	Oxidation	0.43% NO, 2.01% O_2, 1.6% N_2O 7500 hr^{-1}
Zeolites	Khar'k. Politekh. Inst. Im. Lenina	NH_3	0.3% NO, 3-3.8% NH_3, 3-3.8 % O_2 1600 to 10,000 hr^{-1}
Cu + alkaline earth or alkali on Al_2O_3	Hitachi Shipbuilding	CO	530-570 ppm NO, 1.1 - 1.2% CO, 0.4 - 0.5% O_2, 13% CO_2, 10% H_2O, 10,000 hr^{-1}
7.3 Cu/3.5 Ni/89.1 Al_2O_3	Esso Research & Engr	CO, H_2	1400 ppm NO, 1.5% CO, 0.5% H_2, 12% H_2O, 12% CO_2, 0-0.8% O_2, 600 ppm C_4H_{10}, 100,000 hr^{-1}.
SnO_2 and/or CeO_2	Asahi Glass Co.	NH_3	750 ppm NO, 1000 ppm NH_3, 44,000 hr^{-1} 370°C
V-doped Cu-Ni alloys	Texas Instruments	CO/H_2	1700 ppm NO, 500 ppm C_4H_{10} 11% CO_2, 1.2% O_2, 1.5% CO, 1.5% H_2O, 0.5% H_2, 50,000 hr^{-1}, 1175°F.
CuO or Fe_2O_3 (5 - 40%) and Cr_2O_3 (1 - 10%) on Al_2O_3	Agency Ind. Sciences and Technol.	NH_3	800 ppm NO, 1000 ppm NH_3, 4% O_2, 200-350°C
Cu (7.6%) and Co (5.8%) on Al_2O_3	Hitachi Shipbuilding	CO	500 ppm NO, 0.4% O_2, 1% CO, 13% CO_2, 10,000 hr^{-1}, 450°C.
Magnetite (Fe_3O_4) on Al_2O_3	Kanebo Ltd.	CO	2000 ppm NO, 2% CO, 10% CO_2, 11,000 hr^{-1}, 400°C.
Fe oxides	Khar'k Politekh. Inst. Karkov, USSR	NH_3	NO/NH_3 1.15 - 1.25, 6-8% O_2 2-4% H_2O 5000-13,200 hr^{-1}, 250-310°C
CuO (2.8%) on Al_2O_3	Hitachi, Ltd.	NH_3	735 ppm NO, 800 ppm SO_2, 570 ppm NH_3, 1% O_2, 12% H_2O, 12% CO_2 350°C

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
(250°C) 88% oxidation	---	NR	NR	73	
(300°C) 100%	yes	NR	NR	78	Zeolites studied were A-erionites (A = H,Ni, Mg.)
(450°C) 100%	NR	NR	NR	80	
(1100-1400°F) 84-99%	no	NR	NR	87	
15 ppm	NR	NR	NR	91	
> 85 %	NR	NR	NR	111	
> 80%	yes	yes	NR	114	
100%	no	NR	NR	115	
100%	no	NR	NR	124	
50 ppm	yes	NR	NR	129	
100%	yes	yes	NR	130	

(cont.)

TABLE A-2 (cont.)

Catalyst	Company/Institution	Reductant	Operating Conditions
Cu-Ni-Al with Al removed from surface layer	Hitachi Shipbuilding	CO	700 ppm NO, 2% CO, 0.3% O_2 13% CO_2 15,000-20,000 hr^{-1}
Pt group on CeO_2 - impregnated Al_2O_3	Hitachi Makuseru	CO/H_2	1500 ppm NO, 700 ppm HC, 1.5% CO, 1% H_2, 0.5% O_2, 12% CO_2, 3% H_2O,20,000 hr^{-1}
Ni, Fe and Cr (5 - 50%)	Nippon Oil Co.	CO/H_2	3000 ppm NO, 2.0% CO,11.1% H_2O, 0.3% O_2, 0.4% H_2, 30,000 hr^{-1}
$V_xA_yO_z$ (see remarks) on Al_2O_3	Sumimoto Chemical	NH_3	275 ppm NO, 594 ppm NH_3, 11.1 % H_2O,2.6% O_2, 13,000 hr^{-1} 250 - 300°C
Fe_2O_3 - CuO - Ag_2O on Al_2O_3	Asahi Chemical	CO/H_2	300 ppm NO_x, 4.3% CO, 1.6 % O_2, 1000 ppm HC, 20,000 hr^{-1}, 600° C.
CuO (0.5 - 20% as Cu) on Al_2O_3	Hitachi Ltd	NH_3	330 ppm NO_x, 21 ppm SO_2, 1% O_2, 14.1% H_2O, 11% CO_2, 250-500°C.
CuO - Al_2O_3 Synnel (1-10% Cu)	Hitachi Ltd	NH_3	735 ppm NO, 800 ppm SO_2, 570 ppm NH_3, 1% O_2, 12% H_2O, 12% CO_2, 350°C
Pt or Pd - Al_2O_3	Toyoto	CO/H_2	Auto exhaust, 30,000 hr^{-1} 500°C
La-Cu-Zr on Al_2O_3	Hitachi Ltd	NH_3	735 ppm NO, 800 ppm SO_2,570 ppm NH_3m 1% O_2, 12% H_2O,12 % CO_2 > 300°C
Ru and optionally Rh or Pt on Co - ZrO_2	Hitachi Makuseru	CO/H_2	1500 ppm NO, 700 ppm HC, 1.5% CO, 1% H_2, 0.5% O_2, 12% CO_2, 3% H_2O, 20,000 hr^{-1}
Ru and optionally Rh or Pt on Ni - ZiO_2	Hitachi Makuseru	CO/H_2	1500 ppm NO, 700 ppm HC, 1.5% CO, 1% H_2, 0.5% O_2, 12% CO_2, 3% H_2O rest N_2 20,000 hr^{-1}

Conversion or Outlet Conc.	Solution	SO_2 Resistant	Duration	Ref. No.	Remarks
>95 %	no	NR	NR	131	
110 ppm	no	NR	NR	144	
8.9 %	no	NR	81 % at 100 hrs reported	147	Ni:Fe:Cr = (50-90) : (3-40) : (3-40)
96-100%	yes	NR	NR	148	A = Cu, Zn, Sn, Pb, Ti, P, Cr, Fe, Co or Ni; x, y = 0.5-12 z = 1-60 example: $V_9Cr_3O_{27}$
90%	no	NR	"long life"	149	
3.2 ppm	yes	yes	NR	150	Catalyst activated in SO_2 or S-containing solution.
90%	yes	yes	NR	151	
92%	no	NR	NR	153	
99.7%	yes	yes	NR	156	Activated in SO_2 or S-containing solution.
115 ppm	no	NR	NR	157	
110 ppm	no	NR	NR	158	

(cont.)

(cont.)

Catalyst	Company/Institution	Reductant	Operating Conditions
Ru and optionally Rh or Pt on Ni-CeO_2-Zr-O_2	Hitachi Makuseru	CO/H_2	1500 ppm NO, 700 ppm HC, 1.5% CO, 1% H_2, 0.5% O_2, 12% CO_2, 3% H_2O, 20.000 / hr.
Ru and optionally Rh or Pt on Co-CeO_2-ZrO_2	Hitachi Makuseru	CO/H_2	1500 ppm NO, 700 ppm HC, 1.5% CO, 1% H_2, 0.5% O_2, 12% CO_2, 3% H_2O, 20,000/hr.
Metal powder obtained by thermal dissach of metal carbonyl and/or its oxide	Mitsui Mining and Smelting Co. Ltd.	CO	1000 ppm NO_x, 1.7% CO, 1.2 % O_2 1.) 35 x 10^3 hr^{-1} 3.) 20x10^3/hr 2.) 25 x 10^3 hr^{-1}
Ru and Rh on Al_2O_3	Hitachi Ltd.	CO	2500-3000 ppm NO, 500-900HC 2.5-3.0% CO, 0.8-1.0% O_2 350°C.
Pt-Rd	Mitsubishi Heavy Industries	CH_4	CH_4(240 m^3/hr), air (360m^2/hr) mixed with 11.7% O_2, 6.1% CO, 3.9% H_2O 700 ppm SO_2, 1200 ppm NO_x (3000 m^3/hr).
Pt, Pd, Rh or Ni	Sumitomo Chemical Co. Ltd.	H_2, CH_4 C_2H_2, C_4H_x	H_2O (0.5 m^3/hr) added to 0.2% NO_x, 2.1-2.8% O_2(17-18) m^3/hr) and 5-7% H_2, 45-55% CH_4, 15-25% C_2H_4, 1.2% C_2H_4 15-25% C4 components (.25 - .27 m^3/hr, 43-40 vol % .
Ce, U Al_2O_3	Sumitomo Chemical Co. Ltd.	NH_3	2000 ppm NO_x, 3000 ppm NH_3 10 % O_2 (vol) 420°, 40,000 hr^{-1}
Fe-Cr oxide	Katsumata, Tsuyoshi Seo, Tateo	NH_3	72% N_2, 3% O_2, 10% H_2O, 13% CO_2, 250 ppm NO_x, 600 ppm SO_2 and NH_3 (NH_3/NO = 2) 450°, 13,000 hr^{-1}
1.) Cu-Ni-C (4:1:5) 2.) Cu-Ni	Fujikura Cable Works	CO/H	800-950 ppm NO_x, 4-6% CO 300-400 ppm Hydrocarbons .5-.9% O_2. 20,000 hr^{-1}

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
100 ppm	no	NR	NR	159	
90 ppm	no	NR	NR	160	
1.) 65% 2.) 85% 3.) 85%	no	NR	NR	161	
100%	no	NR	NR	162	
30 ppm	yes	NR	NR	163	
50-70 ppm NO_x	no	NR	NR	164	
75.5%	yes	NR	NR	165	Effectiveness increased by treating support with H_2SO_4 or HCl.
80 %	yes	yes	NR	169	
90%	no	NR	1.) ~ 1100 hr 2.) ~250 hr	172	In reduction of NO_x in waste gas, Ni was more effective than Cu.

(cont.)

(cont.) Catalyst	Company/Institution	Reductant	Operating Conditions
Carbonaceous materials (Petroleum coke, coal coke)	Fujikura Cable Works	H_2/Hydrocarbons	Catalyst at 350°,150 ppm NO, 3% H_2, 6ℓ/min.
Ru and Rd	Hitachi Makuseru Co. Ltd.	Hydrocarbons/ CO/H_2	1500 ppm NO,700 ppm hydrocarbons, 1.5% CO, 1% H_2,0.5% O_2, 12% CO_2, 3% H_2O, 20,000 hr^{-1}.
$CoO - \alpha - Fe_2O_3$	Kanebo Ltd.	CO	2% CO, 10% CO_2, 12% H_2O, 20,000 ppm NO, 30,000 hr^{-1}.
Pt impregnated Al_2O_3	Mitsubishi Heavy Industries	CH_4	1% O_2, 10% H_2O,12% CO_2,350 ppm NO,20 ppm SO_2,450° C.
Pt	Environics Inc.	NH_3	100 ppm SO_2, 225 ppm NO, 2% O_2, 450 ppm NH_3.
$V_2O_5 - Al_2O_3$	Khar'k Politekh Inst. im Lenina, Kharkov USSR.	NH_3	.11 vol %NO, .11 vol %NH_3, 10,000 hr^{-1}, 250 - 290°C.
Ni-Ce oxide	Hitachi Maxell Ltd.	CO	0.1% NO, 2% CO, 800°, 20 ℓ/hr.
Pt compounds	Maruenu Seisakusho Co. Ltd.	decomposition	94 ppm NO_x in N_2, 180°C.
Cu, Rh or Ru and alkali + alk. earth or transition metals	Hitachi Shipbuilding and Engineering Co. Ltd.	CO	$[CO]/(2[O_2]+[NO]) = 1.0 - 1.5$, 450°C.
Pt on al.	Markvart, Pour, Koci.	NH_3	30% stoich excess of NH_3, 220 °C, 30,000 hr^{-1}
Cu, La, Zr oxides	Hitachi Ltd.	NH_3	735 ppm NO, 800 ppm SO_2, 570 ppm NH_3, 12% H_2O.12% CO_2, rest N_2, 350°C.
Ru-Pd	Hitachi Ltd	CO/hydrocarbons	3000 ppm NO,500 ppm hydrocarbons, 3% CO, 30,000 hr^{-1}, 400° C.
$Fe_2O_3 - CuO - Cr_2O_3$	Hitachi Ltd.	NH_3	300 ppm NO, 20 ppm NO_2,500 ppm SO_2, 20 ppm SO_3, 250ppm NH_3, 2% O_2 12.5% CO_2, 10% H_2O, 10^4/hr, 300°C.

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
100%	no	NR	NR	175	
40 ppm	no	NR	NR	176	Gas also contained 180
100%	no	NR	NR	177	
95%	no	yes	NR	178	CH_4 / H_2O added to exhaust gas.
90 %	yes	NR	NR	184	
~100%	yes	NR	NR	187	
90-100%	no	NR	1000 hr	192	
94%	no	NR	NR	193	
~100%	no	yes	NR	194	
90%	yes	NR	6 months	196	
99%	yes	yes	NR	197	
> 98%	no	NR	NR	198	
> 90 %	yes	yes	NR	200	(cont.)

(cont.) Catalyst	Company/Instutution	Reductant	Operating Conditions
V_2O_5-MnO_2	Hitachi Ltd.	NH_3	300 ppm NO, 20 ppm NO_2, 500 ppm SO_2, 20 ppm SO_3, 250 ppm NH_3, 2% O_2, 12.5% CO_2, 15% H_2O, 10^4/hr, 300°C.
CuO-Cr_2O_3	Hitachi Ltd.	NH_3	300 ppm NO, 20 ppm NO_2, 500 ppm SO_2, 250 ppm NH_3, 2% O_2, 12.5% CO_2, 15% H_2O, 300° C, 10^4/hr.
MnO_2 and Ni oxide	Toray Industries Inc.	Oxidation	0.43% NO, 2.01% O_2, 1.6% H_2O, 7500 m^3/m^3/hr, 250°C.
NiO, Ru	Mitsubishi Mining Co, Ltd.	CO	1850 ppm NO, 2% CO, 8% O_2, 10% H_2O, 10,000 hr^{-1},
Cu, La, Zr oxides	Hitachi Ltd.	NH_3	735 ppm NO, 800 ppm SO_3, 570 ppm NH_3, 1% O_2, 12% H_2O, 12% CO_2.
Pt group and Ru or Ru oxide	Hitachi Ltd.	CO hydrocarbons	2500-3000 ppm NO, 500-900 ppm hydrocarbons, 0.8% O_2, 2.5-3.0% CO, 20,000 hr^{-1}.
NiO-CuO	Japan Gasoline Co. Ltd.	C_3H_8	800 ppm NO, 500 ppm C_3H_8, 1% CO, 10% H_2O, 12,000 hr^{-1}.
NiO-CuO	Hitachi Ltd.	NH_3	300 ppm NO, 500 ppm SO_2, 20 ppm NO_2, 20 ppm SO_3, 250 ppm NH_3, 2% O_2, 15% H_2O, 12.5% CO_2, 2ℓ/min.
V_2O_5 (10%), OsO_2 (0.1%), CoO (0.8%), Al_2O_3	Mitsubishi Heavy Industries, Ltd.	SO_2	300 ppm NO_x, 500 ppm SO_2, 2% O_2, CO_2 and H_2O present (Baler exhaust), 80,000 hr^{-1}, 400°C.
MnO_2	Toray Industries Inc.	Oxidation	0.43% NO, 20% O_2 (vol), 248°C, 7500 hr^{-1}.
V_2O_5-Cr_2O_3-CuO	Hitachi Ltd.	NH_3	300 ppm NO, 20 ppm NO_2, 500 ppm SO_2, 20 ppm SO_3, 250 ppm NH_3, 2% O_2, 12.5% CO_2, 15% H_2O, 10^4 hr^{-1}, 300°C.

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
>80%	yes	yes	NR	201	
>80%	yes	yes	NR	202	
84-86 %	—	NR	NR	204	80% conversion with O_2 only
99%	yes	NR	NR	205	
>93%	yes	NR	NR	212	
>98%	no	NR	NR	214	
100%	no	NR	NR	216	
93 %	yes	yes	NR	217	
30 ppm	yes	yes	NR	221	SO_2 oxidized to SO_3, 10 ppm residual SO_2.
91%	--	NR	NR	222	
90%	yes	yes	NR	223	

(cont.)

(cont.)

Catalyst	Company/Institution	Reductant	Operating Conditions
V_2O_5, Cr_2O_3	Hitachi Ltd.	NH_3	300 ppm NO, 20 ppm NO_2, 500 ppm SO_2, 20 ppm SO_3, 250 ppm NH_3, 12.5% CO_2, 15% H_2O, 2% O_2, s.v. 10^4/hr, 300°C.
MFe_2O_4 (M=Cu, Ni, Mg, Zn) and/or $(M_{0.5}M'_{0.5})Fe_2O_4$ M, M'=Cu, Ni, Co, Zn)	Kanegafuchi Chem. Ind. Co. Ltd.	CO	3 vol.% CO, 7×10^3 ppm NO. (1) 300°C. (2) 400°C, (3) 500°C, 3.6×10^{-3}, hr^{-1}.
V_2O_5 and CuO	Hitachi Ltd.	NH_3	300 ppm NO, 20 ppm NO_2, 500 ppm SO_2, 20 ppm SO_3, 250 ppm NH_3, 12.5% CO_2, 15% H_2O, 2.5×10^4/hr, 300° C.
CuO (>5 wt %) and MnO_2. MnO_2/CuO M.R. = .12-5.8.	Hitachi Ltd.	NH_3	Same as above. (1) 250°C, (2) 300°C.
Fe_2O_3 and metal oxides-- Al_2O_3.	TDK Electronics Co., Ltd.	CO C_3H_8	1.0% CO_2, 0.1% C_3H_8, 0.1% NO, 0.64% O_2, 10% H_2O, 2×10^4/hr.
LaCoO3 and doped $LaCoO_3$	U.C.L.A.	CO	NO, CO variable concentration, variable temperature.
Pt, Pd, Ru and Rh catalysts	U.C.L.A.	NH_3	14% CO_2, 5% H_2O, 3% O_2, 250-1000 ppm NO, NH_3/NO 0.5-2.4, 10,000 hr^{-1}, 250°C.
Ru, Pd on cordierite	Toyota Central Research and Development Labs, Inc.	CO/HC	Exhaust gas at 400°C.
0.10 - 0.15% Pd and 0.10-0.15% Ru on γ-Al_2O_3	Tr. Mosk. Khim.-tekhnol. Inst. (USSR)	oxidation	0.42-0.4 % NO, 4.8-4.9% O_2, 50-300°C.
Al-Fe alloy layers on Fe and Ru, Ru + Rh or Ru + Pt	Hitachi Maxell, Ltd.	CO/H_2/HC	Automobile exhaust gases, 5×10^4/hr.
Noble metals in Al_2O_3 layers on metals.	Hitachi Maxell, Ltd.	CO	0.1% NO, 2% CO, 20,000 hr^{-1}.

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
~90%	yes	yes	NR	224	High catalytic activity at high temperatures. $V_2O_5/Cr_2O_3 = 0.5-2.0$
(1) 95% (2) 99% (3) 99%	no	NR	NR	225	
>80%	yes	yes	NR	226	$CuO/V_2O_5 = 0.04 - 1.0$ $V_2O_5 > 10$ wt %
(1) 80% (2) 87%	yes	yes	NR	227	
90%	NR	NR	NR	228	10% wt. of a mixt. Fe_2O_3 (40%), MnO_2 (48%) and CuO (12%).
<80%	NR	NR	NR	229	
90%	yes	yes	NR	230	Conversion of NO is a function of NH_3/NO inlet ratio from 0.5-2.3.
86%	no	NR	NR	231	
NR	--	NR	NR	232	
>80%	no	NR	NR	233	
95 ppm	no	NR	NR	234	

(cont.)

(cont.)

Catalyst	Company/Institution	Reductant	Operating Conditions
Pt, Ru and Mg and/or Sr	Toyota Central Research and Development.	CO/HC	Automobile exhaust gases 500^oC, 3×10^4/hr.
$BaRuO_3$ and $MgRuO_3$	Johnson, Mathey and Co., Ltd.	CO	4.0% CO, 2.5% H_2O, 0.4% O_2, 0.2% NO, 0.05% CH_4.
Y zeolite [M-Y] type	Fac. Eng., Kyushu Univ. Fukoka, Japan	NH_3	Not specified in abstract.
Ru on Al_2O_3	Bell Lab., Murray Hill N.J.	CO/H_2	NO, CO, H in the presence of H_2O and CO_2.
CeO_2, α - Al_2O_3	Mitsui Toatso Chemicals, Inc.	NH_3	500 ppm NO_x, 500 ppm NH_3 4 % O_2, 12,000/hr, 350^oC.
V_2O_5 - Al_2O_3 (10%) wt. V_2O_5	Mitsubishi Chemicals, Ind., Co., Ltd.	NH_3	1700 ppm NO. 300 ppm NO_2, 3000 ppm NH_3, 10% O_2, 2×10^4 hr. (1) 300^o (2) 420, (3) 500^oC.
Oxides of Mn, Cu and/or Ag.	Mitsui Toatsu Chemicals, Inc.	NH_3	500 ppm NO_x, 500 ppm NH_3, 4% O_2, 2×10^4/hr, 250^oC.
Oxides of Mn, Cu and/or Ag and $\geq$ 1 % Fe, Co or Ni on carriers.	Mitsui Toatsu Chemicals, Inc.	NH_3	500 ppm NO_x, 500 ppm NH_3, 4 vol. % O_2, 10^4/hr, 300^oC.
Oxides of Mn and $\geq$ 1% of Cr, Mo or W on carriers.	Mitsui Toatsu Chemicals, Inc.	NH_3	500 ppm NO_x, 500 ppm NH_3 4 vol. % O_2, 1.2×10^4/hr, 310^oC.
Oxides of Mn, Cu and/or Ag, and $\geq$ 1% Cr, Mo or W if needed on carrier.	Mitsui Toatsu Chemicals, Inc.	NH_3	500 ppm NO_x, 500 ppm NH_3, 4 vol. % O_2, 2×10^4/hr, 290^oC.

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
87,92,97,98% for air/fuel ratio 13,13.5,14 and 14.5	no	NR	NR	235	
NR	no	NR	NR	236	
NR	NR	NR	NR	237	Catalytic activity was enhanced considerably when a second cation esp., Co(II) or Fe(III) was coexchanged together with Cu(II) in Y-Z colite.
NR	no	NR	NR	238	Ru is active in the formation of CH_4 from CO and H_2.
40 ppm	yes	NR	NR	239	
(1) 76% (2) 90% (3) 87%	yes	NR	NR	240	
5 ppm	yes	NR	NR	241	
5 ppm	yes	NR	NR	242	
10 ppm	yes	NR	NR	243	
5 ppm	yes	NR	NR	244	

(cont.)

(cont.) Catalyst	Company/Institution	Reductant	Operating Conditions
Oxides of Mn and Fe, Co and/or Ni on carriers.	Mitsui Toatsu Chemicals, Inc.	NH_3	500 ppm NO_x, 500 ppm NH_3 4 vol. % O_2, 8000/hr, 300°C.
Fe(> 5 wt % Fe_2O_3) and Cu	Hitachi, Ltd.	NH_3	300 ppm NO, 20 ppm NO_2, 500 ppm SO_2, 20 ppm SO_3, 250 ppm NH_3, 2% O_2, 12.5% CO_2, 15% H_2O, 2000 ml/min 220-490°.
Rare earth oxides	Yamaguchi Univ.	oxidation	NO
Sulfides of Cu, Mn, Ni, Fe and Co	Kuroshiki Boseki K. K.	NH_3	200 ppm NO_x, 1200 ppm SO_2 1% CO_2, 5% O_2, 10% H_2O, 260 ppm NH_3, 500 ml/min, 400°C.
Oxides of rare earth metals (La, Pr, Nd, Sm, Gd) Cu oxide and Zr oxide into Al_2O_3 hydrate.	Hitachi Ltd		550 ppm NO, 940 ppm SO + SO_3, 1.2% O_2, 12.1% H_2O, 12.3% CO_2, 0.435 Kg NH_3/hr, 5 x 10^4/hr, 360°C.
Sintered Ferrite	Kanebo, Ltd	CO	1% CO, 10% CO_2, 12% H_2O, 1000 ppm NO, 30,000/hr. (1) 500, (2) 600, (3) 700 (4) 800°C.
SnO_2, V_2O_5	Asahi Glass Co., Ltd	NH_3	8% O_2, 10% H_2O, 800 ppm SO_2, 750 ppm NO_x, (650 ppm NO) with NH_3 in a mole ratio NH_3/NO_x=1.1, 10,800/hr.
Fe oxide supported on aluminum	Univ. Calif. Berkeley	CO/H_2	Flue Gas
SnO_2 and 1% Cr_2O_3	Garzuakciokinet Tansz, Magy. Tud. Akad., Szeged, Hung.	CO, H_2 C_2H_4	Not specified in abstract .

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
10 ppm	yes	NR	NR	245	
> 90 %	yes	yes	NR	246	FeO_3/CuO mole ratio 1.5 - 20.
NR	--	NR	NR	248	Catalytic activity of nonsesqui oxides > sesqui oxides.
93%	yes	yes	NR	249	
23 ppm	yes	yes	NR	250	
(1) 28 % (2) 85% (3) 100% (4) 100%	no	NR	NR	251	Equal moles of α-Fe_2O_3 and CoO.
NO_x conc. 90, 4, 2, 5, 3.5, 130 ppm at 275, 344, 373 383, 407, 430°C. resp.	yes	yes	NR	252	
"NO and O_2 are reduced"	no	no	NR	253	
NR	NR	NR	NR	254	

(cont.)

TABLE A-2 (cont.)

Catalyst	Company/Institution	Reductant	Operating Conditions
Oxides of Mn and at least one of Fe, CO and Ni and at least one of Cr, Mo and W on carrier.	Hirokawa, Takahashi, Nagayama MITS TOA	NH_3	500 ppm NO_x, 500 ppm NH_3 4% O_2, 10^4/hr, 350°C.
MnO_x, V_2O_3, La_2O_3, CeO_2, Nb_2O_5 and/or Fe_2O_3	Toyota Central Research and Development Laboratories.	CO/HC	NO 1000 ppm (exhaust gas)
NiO, Ru (0.1 - .5 wt. %) Al_2O_3	Mitsubishi Metal Corp.	CO	1800 ppm NO. 1.8% CO, 0.7% O_2, 10% H_2O 10,000/hr, (1) 400, (2) 500 (3) 600°C.
Pt	Stanford University	Decomposition of NO by Pt	NO, O_2
Fe_2O_3 - γ- Al_2O_3	Tokyo Inst. Technol.	H_2	Not specified in abstract
Ru (0.02 - 0.85%) Pt, Pd and/or Rh (0.001 - .7 %)	Nissan Motor Co. Ltd.	CO/H_2	1000 ppm NO, 500 ppm propane, 2.0% CO, 12.0% CO_2, 5% O_2, 1.0% H_2, 5.0 % H_2O 290°C.
Cu and ≥ 1% of alkali, alk. earth, transition or noble metals on γ- alumina support.	Hitachi Shipbuilding.	NH_3	4.2% O_2, 12% CO_2, 10% H_2O, 350 ppm NO, 200 ppm SO_2, 520 ppm NH_3, 2500/hr (1) 240, (2) 290 (3) 340 (4) 380°C.
$M_2O \cdot Fe_2O_3$ (M = Li, Na, K, Ba, Cu)	Kanegafuchi Chem. Industry	CO	3% CO, 4500 ppm NO. 3900/hr. (1) 250 (2) 300 (3) 400°C.
Fe oxide and V oxide	Asahi Glass Co., Ltd	NH_3	8% O_2, 10 % H_2O, 800 ppm SO_2, 750 ppm NO_x, 650 ppm NO, NH_3/NO_x mole ratio = 1.1, 10,800/hr
Fe oxide and Ce and/or Bi oxide(s)	Asahi Glass Co., Ltd	NH_3	8% O_2, 10% H_2O, 750 ppm NO_x, (650 ppm NO), NH_3/ NO_x mole ratio = 1.1. 10,800/hr.

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks.
5 ppm	yes	NR	NR	255	
150 ppm	no	NR	NR	256	
(1) 85 % (2) 98% (3) 100%	no	NR	NR	257	
NR	yes	NR	NR	259	
NR	NR	NR	NR	260	
90 %	no	NR	NR	261	
(1) 83% (2) 100 % (3) 98% (4) 99%	yes	yes	NR	262	Catalyst tested: Rh (0.03% Cu (5 %) and Mg (3%)
(1) 65% (2) 98% (3) 100%	no	NR	NR	263	
NO_x: 200, 10, 35, 13, 7, 20, 100, 200 ppm at 260, 285, 302, 335, 370, 400, 420, 450°C, resp.	yes	NR	NR	265	
NO_x: 10, 50, 18, 1, 5 ppm at 305, 320, 343 395, 427°C resp.	yes	NR	NR	265.	

(cont.)

(cont.) Catalyst	Company/Institution	Reductant	Operating Conditions
Cu (0.1-10%), La (0.1- 20%), Zr (0.01-12%) oxides or double oxides	Hitachi, Ltd.	CO/HC	Exhaust gas pipe of a 4-cylinder gas engine.
0-contg. S compd. and Fe, Cu, Cr, Mn, V, Ni and/or Ti	Hitachi, Ltd.	NH_3	300 ppm NO, 20 ppm NO_2, 500 ppm SO_2, 20 ppm SO_3 250 ppm NH_3, 12.5 %O_2, CO_2, 15% H_2O (vapor), 5 x 10^4/hr, 300°C.
SnO_2, MoO_3	Asahi Glass Co. Ltd.	NH_3	750 ppm NO_x, (650 ppm NO) with NH_3 at mole ratio NH_3/NO_x = 1.3. 10,000 hr^{-1}, 450°C.
Cu-Al Alloys	Mitsui Toatsu Chemicals	NH_3	500 ppm NH_3,500 ppm NO_x, 4% O_2, 10,000 hr^{-1}, 200-500° C.
$V_9Cr_3O_{27}$	Sumitomo Chemical Co., Ltd.	NH3	427 ppm NO, 613 ppm NH_3 2.8 % O_2, 89.4 % N_2,7.8%, H_2O, 15,600 hr^{-1}.
$Cr_xA_yO_z$	Sumitomo Chemical Co., Ltd.	NH_3	2.6% O_2, 11.7% H_2O. 307 ppm NO, 629 ppm NH_3, 1.36 x 10^{-4}/hr, 200° - 250°.
V_2O_5/Fe_2O_3	Hitachi Ltd.	NH_3	300 ppm NO, 20 ppm NO_2, 500 ppm SO_2, 20 ppm SO_3, 250 ppm NH_3, 2 % O_2,12.5% CO_2, 15% H_2O, 10^4 hr^{-1}, 250-450°C.
Cu, Ni, Ir, Ru	Japan Catalytic Chemical Industry Co., Ltd.	NH_3	1000 ppm NO_x, 2.5 ℓ/min. (1) 300, (2) 450 (3) 600 (4) 750°C.
Pt group	Sumitomo Chemical Co.	H. CH_4, C_2H_6, etc.	0.5 m^3/hr H_2O, 0.2% NO_x, 2.1-2.8% O_2, 17-18 m^3/hr, 430-440°C.
Ni-Cu-Cr	Kobe Steel Ltd.	CO	2.7% NO, 7.7% CO, 2000/hr, 650°C.

Conversion Outlet Conc.	Selective	SO_2 Resistant	Duration	No.	Remarks
~99%	no	NR	NR	266	Porous ceramic carriers.
> 80%	yes	NR	NR	267	
45 ppm	yes	NR	NR	268	NO_2= 0 ppm.
> 95%	yes	NR	NR	270	
9 ppm	yes	NR	NR	271	NH_3 3 ppm.
> 91%	yes	NR	NR	272	A = Ni, CO, Zn, Mo, W, Sn, P, Ti.
~85%	yes	yes	"improved durability"	273	x, y = 0.5 - 12% z = 1 - 54%.
(1) 82%, (2) 92% (3) 94%, (4) 95%	NR	NR	NR	274	
50 - 70 ppm	no	NR	> 70 hr	278	
100 %	no	NR	NR	280	

(cont.)

TABLE A-2 (cont.)

Catalyst	Company/Institution	Reductant	Operating Conditions
CeO_2 — Fe oxide	Mitsubishi Chemical Ind. Co., Ltd.	NH_3	2000 ppm NO_x (300 ppm (NO), 3000 ppm NH_3, 10% vol O_2, 2 x 10^4/hr, (1) 350, (2) 410, (3) 440°C.
Fe oxide	Mitsubishi Chemical Ind. Co. Ltd.	NH_3	10% O_2, 1700 ppm NO, 300 ppm NO_2, 3000 ppm NH_3, 2 x 10^4/ hr^{-1}, (1) 350 (2) 400 (3) 480° C.
Ce oxide	Mitsubishi Chemical Ind. Co. Ltd.	NH_3	1700 ppm NO, 300 ppm NO_2 3000 ppm NH_3, 10% O_2, 2 x 10^4 hr^{-1}, (1) 360, (2) 390 (3) 420 (4) 460° C.
Rh, Ru	Asahi Chemical Industry Co. Ltd.	CO	2.2 - 2.5 % CO, 2% O_2, 350 ppm NO_x, 3.2 x 10^4 hr^{-1}. (1) 350, (2) 500°C.
Cu	Research Foundation for the Development of Industries.	NH_3	620 ppm NO, NH_3/NO=1.53 1.2 x 10^4 hr^{-1}, 305°C.
Zeolite with Cu	Toray Industries Inc.	NH_3	0.3% NO, 3% NH_3, 5% O_2, 1.6% H_2O, 10,000 hr^{-1} 400°C.
Ce, W	Hitachi, Ltd.	NH_3	720 ppm NO, 910 ppm NH_3 4% O_2, 21,000 hr^{-1}, (1) 300 (2) 350, (3) 400 (4) 450 (5) 500°C.
SiO_2, Si acid or silicates	Kobe Steel, Ltd.	oxidation with HNO_3	200 ppm NO, 3% O_2, 400 ppm HNO_3, 100% rel. humidity at 27°C.
Pt	Mitsubishi Heavy Ind. Ltd.	NH_3	11% CO_2, 15% H_2O, 2.5% O_2, 175 ppm NO_x, 3200 ppm SO_2, 176 ppm NH_3.
Fe compound	Hitachi Ltd.	NH_3	200 ppm NO_x, 500 ppm SO_2, 20 ppm SO_3, 40 mg/m^3 dust, 10,000 hr^{-1}, 350°C.

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
(1) 60.9% (2) 98.2% (3) 64.9%	yes	NR	NR	281	
(1) 77.5% (2) 87.3% (3) 90.5% (4) 92.3%	yes	NR	NR	282	
(1) 69.5% (2) 73.75% (3) 76.25% (4) 73.0%	yes	NR	NR	283	
NO_x: (1) 85% (2) 98% Ru: (1) 82% (2) 95% (Ru).	no	NR	NR	288	
97%	yes	NR	NR	289	
99%	yes	NR	NR	291	
(1) 98% (2) 100% (3) 100% (4) 100% (5) 91%	yes	NR	NR	290	
50 - 98%, depending on silaceous catalyst	yes	NR	NR	292	
< 2 ppm	yes	yes	NR	293	
90%	yes	yes	78% after 1000 hrs	299	

(cont.)

(cont.) Catalyst	Company/Institution	Reductant	Operating Conditions
Ir-Al_2O_3 plus Ru-Al_2O_3	Toa Nenryo Kogyo K.K.	C_3H_6 CO, H_2	1500 ppm NO, 400 ppm C_3H_6, 1.0% CO, 10% CO_2, 0.5% H_2, 10% H_2O, (O_2 used/O_2 needed to oxidize all CO, C_3H_6 and H_2)= (1) 0.45, (2) 0.8 (3) 1.0 (4) 1.2 (5) 1.6; 300 m^3/hr, 500°C.
Sulfate of Mn, Cr, Cu, Fe, V or Ce	Mitsubishi Gas Chemical Co., Inc.	NH_3	2400 ppm NO, 10 ppm NO_2, 300 ppm NH_3, 600 ppm SO_2, 6000 hr^{-1}, 250°C.
Na, K, Mg, Al, Sn, Mn, Cr, Ni, Cd, Cu, Fe, Li and/or Be Sulfates plus oxides of V, Mn, Cr, Cu and/or Fe on conventional carriers	Mitsubishi Gas Chemical Co., Inc.	NH_3	240 ppm NO, 10 ppm NO_2, 300 ppm NH_3, 600 ppm SO_2, 3% O_2, 12% CO_2, 6000 hr^{-1}, 250°C.
Sn, W and Mo oxides	Hitachi, Ltd	NH_3	290-310 ppm NO, 240-260 ppm NH_3, 450-550 ppm SO_2 2-4% O_2, 12.5% CO_2, 15% H_2O, 11,000 hr^{-1}, (1) 250 (2) 300 (3) 350 (4) 400 (5) 450, (6) 500°C.
Rh-Al_2O_3	Monsanto Co.	H_2, CO	0.15% NO, 1% CO, 0.33% H_2 ~3% H_2O, 10,000 hr^{-1}, 269°C
V-oxide W-oxide	Sumitomo Chemical Co., Ltd.	NH_3	2.6% O_2, 12.1% H_2O, 555 ppm NO, 617 ppm NH_3, 13,700 hr^{-1}, 250-400 °C.
Fe_2O_3 (94.5%) Cr_2O_3 (5.5%)	Hitachi Ltd.	Oxidation	Not specified in abstract.
Fe, Ag or their oxides with Cu, Pb, In, Ce, Be or their oxides on Al_2O_3.	Asahi Chem., Co.	C_4H_{10}	450 ppm NO_x, 2.5% O_2, (15 m^3/hr) mixed with C_4H_{10} (0.1 m^3/hr).

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
(1) 92% (2) 100% (3) 96% (4) 85% (5) 60%	no	NR	NR	301	Two stage reactor (Ir section first).
96%	yes	yes	NR	306	
18 ppm	yes	yes	NR	307	
(1) 64% (2) 84% (3) 91% (4) 93% (5) 94% (6) 93%	yes	yes	NR	310	Pressed Catalyst
50%	no	NR	NR	311	
>97%	yes	NR	NR	315	V/W = (7-11.7):(0.3-5) molar
NR	--	yes	no deterioration in 100 hrs.	312	
91%	no	NR	NR	316	

(cont.)

(cont.) Catalyst	Company/Institution	Reductant	Operating Conditions
Ru, Pd on Al_2O_3	Hitachi, Ltd.	NH_3	300 ppm NO, 10-15 ppm NO_2 450-500 ppm SO_2, 250-300 ppm NH_3, 3-5% O_2, 10-13% H_2O, 3 x 10^4 hr^{-1}, 350°C.
Cu_2O on stainless steel wire	Fujikura Cable Works Ltd.	CO	1500 ppm NO, 2.1% CO, 12.0% H_2O, 0.5% O_2, 3 x 10^4 hr^{-1} 480°C.
CuO, CoO on Al_2O_3	Mitsui Petrochemical Industries, Ltd.	NH_3	300 ppm NO, 3% O_2, 15% CO_2, 12% H_2O, 400 ppm NH_3, 1.5 x 10^4 hr^{-1}, 350°C.
$CuSO_4$, $FeSO_4$ on Al_2O_3	Okayama University, Okayama, Japan	NH_3	500 ppm NO, 1000 ppm SO_2, 667 ppm NH_3, 15% O_2, 10% H_2O, 1000 N Cm^3/min, <~ 400°C.
$CuSO_4$ on Al_2O_3	Mitsui Petrochemical Industries, Ltd.	NH_3	300 ppm NO, 400 ppm NH_3 3% O_2, 15% CO_2, 12% H_2O, 1.5 x 10^4 hr^{-1}, 350°C.
NiO	Dep. Catalisis, Cons. Super. Invest. Cient. Spain	H_2	0.3% NO_x, 3% O_2, 7% H_2, 372-1969 ml/min, 210-449°C
Mo	Osaka Munic. Tech. Res. Inst., Osaka, Japan.	NH_3	850-950 ppm SO_2, 1100 ppm NO, 1380 NH_3, 5.22-5.31% O_2, 10% H_2O, 10,700 - 900 hr^{-1}, (1) 200 (2) 350°C.
Cu-Cr oxide	Mitsui Toatsu Chemicals Inc.	NH_3	300 ppm NO_x, 50 ppm SO_x, 38 ppm CO, 10% O_2, 6.5% CO_2, 12% H_2O, 30 ℓ/hr NH_3
Ir-Au on Al_2O_3	Toa Nenryo Kogyo K.K.	CO, H_2 propylene	1500 ppm NO_x, 400 ppm propylene, 1.0% CO, 10% CO_2, 0.5% H_2, 10% H_2O, 0.42-15 % O_2, 3 x 10^4 hr^{-1}.
Mn-Cr oxides on Al_2O_3	Mitsubishi Chemical Industries, Co., Ltd.	NH_3	2000 ppm NO_x (300 ppm NO) 3000 ppm NH_3, 10% O_2, 2 x 10^4 hr^{-1}, 320°C.

Conversion or Outlet Conc.	Selective	SO_2 Resistance	Duration	Ref. No.	Remarks
80%	yes	yes	continuous over 100 hr	321	Not readily inactivated by SO_x, O_2, CO_2
90%	no	NR	NR	323	Catalyst with only Cu required 820° for 90% removal.
98%	yes	yes	NR	324	
"Excellent conversion"	yes	yes	NR	328	For $CuSO_4$, presence of SO_x made rate higher.
12 ppm	yes	yes	NR	336	
99%	no	NR	NR	338	Catalyst lost activity but regenerated by heating in O_2 flow.
NO_2 (1) 25.8 ppm (2) 0 ppm	yes	yes	no decrease after 30 hr.	339	
30 ppm	yes	NR	NR	343	
73%	no	NR	NR	344	
72%	yes	NR	NR	345	

(cont.)

(cont.) Catalyst	Company/Institution	Reductant	Operating Conditions
FeCr oxides	Ube industries, Ltd.	NH_3	3% O_2, 1000 ppm NO, 1100 ppm NH_3, 700 ppm SO_2, 15,000 hr^{-1}, 400°C.
Cr_2O_3 on Al_2O_3	Toa Nenryo Kogyo K.K.	CO, H_2 propylene	1500 ppm NO_x, 400 ppm propylene, 10% CO, 10% CO_2, 0.5% H_2, 10% H_2O, 0.42%-1.5% O_2.
Compounds of Ti, Cr, Mn, Fe, Co, Ni, Cu, V, Mo, or W with H_3Bo_3 or H_3PO_4 or their salts on C	Takeda Chemical Industries, Ltd.	NH_3	0.2% NO, 0.3% NH_3, 1% H_2O, 5% O_2, 93.5% He. (1) 110°, (2) 150°, (3) 250°C.
V_2O_5 on γ-alumina	Mitsubishi Heavy Industries, Ltd.	NH_3	8.5% CO_2, 16.8% H_2O, 2.6% O_2, 169 ppm NO_x, 160 ppm NO, 217 ppm NH_3, 10^4 hr^{-1}, 300-400°C.
Rh on Al_2O_3	Mitsubishi Heavy Industries, Ltd.	CO	1500 ppm NO_x, 30 ppm SO_2, 2% CO, 14% CO_2, 10% H_2O, 1% O_2, 300°C.
MoO_3, CoO	Toyo Engineering Corp.	NH_3	600 ppm NO, 50 ppm NO_2, 3% O_2, 6% H_2O, 15% CO_2, 1500 ppm SO_2, plus NH_3 ($NH_3/NO_x = 1.35$) 15,000 hr^{-1}, 500°C.
Fe-W	Mitsubishi Petrochemical Co., Ltd	NH_3	700-900 ppm NO, 850-1100 ppm NH_3, 4% O_2, 1.07 x 10^4 hr^{-1}, 200°C.
V_2O_5 on Al_2O_3	Agency of Industrial Sciences and Technology	NH_3	300 ppm NO, 500 ppm NH_3 3.0% O_2, 9.2% H_2O, 16,000 hr^{-1}. (1) 250°, (2) 300°, (3) 350°, (4) 450°C.
Fe-Al Co-Mn oxides, Pt group	Hitachi Maxell, Ltd.	CO, H_2	1.2% CO, 0.5% H_2, 14% CO_2, 0.3% O_2, 12% H_2O, 0.05% C_3H_8, 0.1% NO, 2 x 10^4 hr^{-1}, 500°C.

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
93%	yes	yes	more than 80 hrs	346	
90%	no	NR	NR	347	
(1) 91% (2) 99% (3) 100%	yes	NR	NR	348	Example Catalyst: $CuSO_4$ on C.
90%	yes	NR	NR	349	
~100%	no	yes	NR	350	Produced ~400 ppm NH_3
110 ppm	yes	NR	NR	351	
99%	yes	NR	NR	352	Fe/W ratio 9/2
(1) 92% (2) 99% (3) 100% (4) 100%	yes	NR	NR	353	
99%	no	NR	NR	356	

(cont.)

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Catalyst	Company/Institution	Reductant	Operating Conditions
Ru-Rh	Hitachi Maxell, Ltd.	CO, C_3H_6/ H_2	1.2% CO, 14% CO_2, 0.05% C_3H_6, 0.3 - 1.0% O_2, 0.6% H_2, 10% H_2O, 0.1% NO
Refractory inorg. oxides coated with CuO and NiO	Ube Industries, Ltd.	NH_3	1660 ppm NO, 2.5% O_2, 1570 ppm NH_3, 7% H_2O, 1.5×10^4/hr, 300°C.
Pt on surface of a conveyor belt.	Inoue Japan Research Institute	NR	Flue gas containing NO_x.
1st layer: NiO-CuO, Nb_2O_3-Y_2O_3-CuO, Nb_2O_3-CoO, Y_2O_3-CoO, CeO_2-, La_2O_3-Fe_2O_3-, Nb_2O_3 - or NiO-Mn_2O_3. See remarks.	Toyota Central Research and Development Laboratories, Inc.	Oxidation	Automobile Exhaust Gases
Pt, Ru, Pd on Al_2O_3	Nippon Shokubai Kagaku Kogyo Co., Ltd.	Oxidation	Waste gas 500°C.
VO_x, Al_2O_3 and BN, Al hydroxychloride	Kanegafuchi Chemical Industry Co., Ltd.	NH_3	400 ppm NO_2, 40 ppm NH_3, 1500 ppm SO_2, 2% O_2, 10% steam, 300-450°C.
Al, Cu, Mn	Hitachi Shipbuilding and Engineering Co., Ltd	NH_3	Exhaust gases
Fe-Cr-Oxides.	Ube Industries, Ltd.	NH_3	1000 ppm NO, 1100 ppm NH_3 3% O_2, 350°C.
NiO, CoO on Al_2O_3	Fujimi Kenmazai Kogyo K.K.	C_3H_8	2% O_2, 0.36% C_3H_8, 13.0% CO_2, 10.0% H_2O, 0.03 vol % NO, 4×10^3/ hr. 500°C.
Mordenite, Fe ions	Toray Industries, Inc.	NH_3	300 ppm NO, 300 ppm SO_2 300 ppm NH_3, 9% H_2O, 2 wt % O_2, 1000 ml/min, 350°C
V_2O_5, MnO, γ-Al_2O_3 carrier	Mitsubishi Chemical Industries Co., Ltd.	NH_3	Waste gas (1) 220°C, (2) 280°C, (3) 340°C.
Cu, Ni, Ir and Rh and/or Pd and Pt	Japan Catalytic Chemical Industry Co., Ltd.	CO/H_2	0.5% CO, 0.44-0.66% O_2, 330 ppm C_3H_6, 500 ppm NO, 0.33% H_2, 10% H_2O, 10^5/hr, 750°C.

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
~200 ppm	no	NR	NR	357	NH_3 0-100 ppm
85%	yes	NR	NR	358	
<1%	NR	NR	NR	359	
80-100%	NR	NR	NR	360	2nd layer: Pt, Pt-Rh, Pt-Pd on NiO-Cr_2O_3-CuO-Fe_2O_3-BaO. Also a layer of Cu, CuO.
92%	NR	NR	NR	361	
90-95%	yes	yes	NR	366	
~60-70%	yes	yes	NR	367	
83.7%	yes	yes	NR	368	
0.0054 vol.%	see remarks	NR	30 min.	370	O_2 is in slight excess.
99.8%	yes	yes	NR	371	
(1) 61.5% (2) 80.3% (3) 84.5%	yes	NR	NR	372	V_2O_5/MnO < 1
93%	NO	NR	NR	373	

(cont.)

(cont.) Catalyst	Company/Institution	Reductant	Operating Conditions
Fe_2O_3, MnO_2, CuO, Ru on Al_2O_3	TDK Electronics Co., Ltd.	NH_3	4% O_2, 400 ppm NO_x, 1000 ppm SO_2, 10% H_2O, 400 ppm NH_3, 5 x 10^3/hr, 400°C.
Ti, V	Mitsubishi Petrochemical Co., Ltd	NH_3	700- 800 ppm NO, 600-700 ppm NH_3, 482 ppm SO_2. 4% O_2, 7.14% H_2O, 5 x 10^4/hr, 250°C.
Ti, V oxides and/or Mo, W, P and/or B oxides	Takeda Chemical Industries, Ltd.	NH_3	0.03% NO, 0.06% SO_2, 6.0% O_2, 10% H_2O, 0.03 vol % NH_3, 5 x 10^3/hr, 200°C.
CuO MnO_2	Mitsui Toatsu Chemicals Inc.	NH_3	(1) 500 ppm NO_x, 500 ppm NH_3, 4% O_2, 15% H_2O, 2 x 10^4/hr, 250°C. (2) same as 1 except O_2 = 8% .
MnO_2, Fe_2O_3, CuO, Ru	TDK Electronics Co., Ltd.	CO	1% CO, 0.1% hydrocarbons, 0.1% NO_x, 10% H_2O, 1% O_2, 10^4/hr, 400°C.
MCo_2O_4	Mitsui Toatsu Chemical, Inc.	NH_3	150 ppm NO, 15 ppm N_2O, 4% O_2. 23% H_2O, 50 ppm SO_2, 10% CO_2, 165 ppm NH_3, 10^4/hr, 300°C.
SnO_2, Cr_2O_3	Asahi Glass Co., Ltd	NH_3	750 ppm NO_x, 800 ppm SO_2, 8 % O_2, 8% H_2O (NH_3/NO_x mole ratio = 1.3) 10,000/hr.
Cu with alkali metals or transition metals and with Rh or Ru on Al_2O_3.	Hitachi Shipbuilding and Engineering Co., Ltd.	H_2	600 ppm SO_2, 500 ppm NO, 0.4% O_2, 1.1% H_2, 15% CO_2, 10% H_2O, 10^4/hr, 450°C.
Sulfates of Co, Mn, Ce and/or Ni, coated with Ru on Al_2O_3	Hitachi Makuseru Co., Ltd.	H_2	Exhaust gases, 2 x 10^4/hr, 1500 ppm NO.
Al alloy, Fe, Ni and Ce oxides	Hitachi Makuseru Co., Ltd.	CO/H_2 HC	Automobile exhaust gases, 5 x 10^4/hr.

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
> 80%	yes	yes	NR	374	
97%	yes	yes	NR	375	
77%	yes	yes	NR	376	
(1) 50 ppm (2) 10 ppm	yes	NR	NR	377	
> 80%	no	NR	NR	378	
83.2%	yes	yes	NR	379	M = Ni, Zn, Mg, Ca, Mn, Cu or Fe.
100%	yes	yes	NR	381	
100%	no	yes	NR	385	
100 ppm	NR	yes	NR	386	
80%	NR	NR	1150 hr	387	

(cont.)

(cont.) Catalyst	Company/Institution	Reductant	Operating Conditions
Carboxylates of Fe metals or Cu on Al_2O_3	Mitsui Mining and Smelting Co., Ltd.	NH_3	Exhaust gas, 1000 ppm NO_x
Ru and Cu, Zn, Ni, Co, Cr and/or Ag.	Hitachi, Ltd.	CO	3% CO, 3000 ppm NO, 3 x 10^4/hr, 300°C.
Cu or CuO on $\gamma - Al_2O_3$	Japan Gasoline Co., Ltd.	NH_3	1100 ppm SO_2, 500 ppm NO 550 ppm NH_3, 3% O_2, 10% H_2O, 94.1 ℓ/hr, 400°C.
SiO_2 -Al_2O_3 coated with Cr, Mn, Fe, Co, Ni and/or Cu oxide	Mitsubishi Chemical Industries Co., Ltd.	NH_3	10% O_2, 2000 ppm NO_x 3000 ppm NH_3, 2 x 10^4/hr, 360°C.
Cr, Mn, Fe, Co, Ni and/or Cu oxides on acid-treated carriers.	Mitsubishi Chemical Industries Co., Ltd.	NH_3	10% O_2, 2000 ppm NO_x, 3000 ppm NH_3, 2 x 10^4/hr, 350°C.
Fe and Cr oxides γ- Al_2O_3 carriers	Mitsubishi Chemican Industries Co., Ltd.	NH_3	Waste gas, 380°C.
Ni, Cu, Cr oxides	Toyota Central Research and Development Lab.	CO/H_2/ HC	Exhaust gas. 600°C, 6 x 10^4/hr.
Alloy of metals from groups I B, III, VII B and VIII	International Harvester Co.	CO/H_2 HC	Exhaust (automobile) 800-1400 $^{\circ}$F.
Si, Si oxide	Kobe Steel, Ltd.	NH_3	370 ppm HNO_3, 0.12% SO_2 210 ppm NO_x [10 ppm NO_2 200 ppm NO], 4% O_2, 12% CO_2.
Ti and ≥ 1% of Mo, W, Fe, V, Ni, Co, Cu, Cr, U and Sn as oxides.	Mitsubishi Petrochemical Co., Ltd., Hitachi Ltd., Babcock-Hitachi K.K.	NR	200-500°C.
Cu alloy containing Zn, Fe, Mg, etc.	Inone Japan Research Inst.	CO/H_2 HC	Waste gas, 3600 ppm NO_2 100 ml/min.

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
78%	yes	yes	NR	388	
40%	NR	NR	NR	389	
90%	yes	yes	NR	394	
75.3%	yes	NR	NR	395	
76.5%	yes	NR	NR	396	Example: γ- Al_2O_3 carrier.
74%	yes	NR	NR	397	
76%	NR	NR	NR	398	Catalyst contains 2-12% O_2, honeycomb shape.
>90%	NR	NR	NR	400	Ni, Cu, Fe, Mn, Y
<5ppm	yes	yes	NR	402	
~95%	NR	NR	NR	404	Example: Ti + Mo (Ti/Mo = 9:1)
13 ppm NO_2	NR	NR	NR	407	(cont.)

(cont.) Catalyst	Company/Institution	Reductant	Operating Conditions
Cu, Al, Fe and SiO_2	Japan Gasoline Co., Ltd.	NH_3	500 ppm NO, 550 ppm NH_3 1500 ppm SO_2, 3% O_2, 10% H_2O.
CuO on Al_2O_3	Fujikura Cable Works Ltd.	Oxidation	1200 ppm NO, 10% O_2, 5ℓ/min.
Ni-Cu-Co (30:10:10)	Nissan Motor Co., Ltd.	NR	(1) 400°, (2) 900° (3) 1200°C.
Ti, Mo, Ni and Fe (Pelletized with C and fired)	Hitachi, Ltd.	NH_3	15% N_2O, 20% H_2, 3% O_2, 5000/hr, 375-500°C.
SiO_2	Kobe Steel, Ltd.	Oxidation with HNO_3	200 ppm NO, 3% O_2, 400 ppm HNO_3.
CeO_2, TiO_2 on α - Al_2O_3	Mitsubishi Chemical Industries Co., Ltd.	NH_3	10% O_2, 300 ppm NO_2, 1700 ppm NO, 3000 ppm NH_3, 2×10^4/hr. (1) 260° (2) 310°, (3) 410°, (4) 450°, (5) 505°C.
CeO_2 on α - Al_2O_3	Mitsubishi Chemical Industries Co., Ltd.	NH_3	10% O_2, 300 ppm NO_2, 1700 ppm NO, 3000 ppm NH_3, 4×10^4/hr. (1) 250°, (2) 310°, (3) 370°, (4) 430°, (5) 460°C.
CeO_2 on α - Al_2O_3	Mitsubishi Chemical Industries Co., Ltd.	NH_3	10% O_2, 300 ppm NO_2, 1700 ppm NO, 3000 ppm NH_3, 4×10^4/hr. (1) 300° (2) 380° (3) 420°C.
Pt on α - Al_2O_3	Mitsubishi Chemical Industries Co., Ltd.	NH_3	10% O_2, 300 ppm NO_2, 1700 ppm NO, 3000 ppm NH_3, 4×10^4/hr. (1) 225° (2) 230°, (3) 235°, (4) 240°C
V-oxide on α - Al_2O_3	Mitsubishi Chemical Industries Co., Ltd.	NH_3	10% O_2, 300 ppm NO_2, 1700 ppm NO, 3000 ppm NH_3, 40,000/hr. (1) 210° (2) 270°, (3) 330°, (4) 360°C

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
NR	yes	yes	NR	413	Catalyst prepared from sulfates, roasted at 550°C.
69%	--	NR	NR	414	
(1) 98% (2) 97% (3) 97%	NR	NR	NR	415	
100%	yes	NR	NR	417	> 5% Ti. Example: Ti:Fe = 9:1. Nitrous oxide reduction.
80% converted to NO_2	NR	yes	NR	419	N_2O subsequently absorbed in 2% $Mg(OH)_2$ slurry.
(1) 54.8% (2) 71.5% (3) 86.3% (4) 87.0% (5) 84.0%	yes	NR	NR	421	
(1) 24.0% (2) 49.0% (3) 73.8% (4) 80.5% (5) 78.0%	yes	NR	NR	422	
(1) 47.5% (2) 72.8% (3) 75.5%	yes	NR	NR	423	
(1) 82.0% (2) 84.0% (3) 84.0% (4) 79.0%	yes	NR	NR	424	
(1) 40.8% (2) 67.0% (3) 81.0% (4) 77.0%	yes	NR	NR	425	

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(cont.) Catalyst	Company/Institution	Reductant	Operating Conditions
V_2O_5	Duengemittel-Prod., BASF, Ludwigshafen Ger.	NH_3	Effluent gases from a HNO_3 plant. 500-1000 ppm NO_x
Cu oxide and alkali metal oxide C-carrier.	Toray Industries, Inc.	NR	0.47 % NO, 6000/hr.
$FeSO_4$ on Al_2O_3	Hitachi Shipbuilding and Engineering Co., Ltd.	Decomposition or Oxidation	500 ppm NO, 500 ppm NO_2, 4% O_2, 12% CO_2, 10% H_2O, 2 x 10^4/hr, 390-500°C.
$CuSO_4$, $FeSO_4$ on γ-Al_2O_3	Mitsui Petrochemical Industries, Ltd.	NH_3	3% O_2, 15% CO_2, 12% H_2O, 300 ppm NO, 1500 ppm SO_2, 200 ppm NH_3, 15,000/hr 350°C.
Fe-containing ores cinders, scales, etc.	Nippon Kokan K.K.	NH_3	7% CO_2, 13% O_2, 79.7% N_2, 0.3% CO, 15 ppm NO_2, 135-155 ppm NO, 200 ppm NH_3. 300 ℓ/hr, 300 - 370°C.
Fe_2O_3 Sn, Sn/Fe at. ratio: 0.01-20 Graphite carrier.	Hitachi, Ltd.	NH_3	290-310 ppm NO, 240-260 ppm NH_3, 450-550 ppm SO_2, 2-4% O_2, 12.5% CO_2, 15% H_2O, 59,000/hr. (1) 300° (2) 350°, (3) 400° (4) 450°C.
Ir on Al_2O_3	Corp. Res. Lab., Exxon Res. and Eng. Co., Linden, N.J.	CO	CO-NO + 75% excess oxidants.
Ni and Thoria, Mullite carrier	Ube Industries, Ltd.	CO	Helium contg.: 0.3% NO, 1.3% H_2, 0.7% O_2, 0.05% C_2H_4, 2.5% CO, 8% CO_2, 5% H_2O, 1800 ppm SO_2, 700°C 5 x 10^4/hr. (1) 0 hr, (2) 1 hr, (3) 2 hr, (4) 5 hr.
ABO_3 Al_2O_3 carrier	duPont de Nemours, E. I., and Co.	CO	2000 ppm NO, 10,000 ppm CO, (1) 385° (2) 450°, (3) 540°C, 40,000/hr,

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
50-150 ppm	yes	NR	NR	426	
99%	NR	NR	NR	428	0.1 - 2.0% Cu. Probable reaction with C.
95%	--	yes	NR	429	
93%	yes	yes	NR	430	Example: $CuSO_4$ 60% SO_4 1.5%
87%	yes	NR	NR	431	
(1) 82.8% (2) 91.5% (3) 97.0% (4) 92.3%	yes	yes	NR	432	Fe $\geq$ 2% (as Fe_2O_3) Sn/Fe = 0.01-20.
90%	yes	NR	NR	435	
(1) 96% (2) 93% (3) 93% (4) 92%	no	yes	NR	436	Example: Ni:Th = 1:0.1
(1) 25% (2) 50% (3) 90%	no	NR	NR	438	A = metal with atomic # 11-51, 56-71, 89-103. B= part Pt-group, part non-Pt ionic radius 0.4-1.4A, valence 3. (cont.)

(cont.) Catalyst	Company/Institution	Reductant	Operating Conditions
ABO_3 Al_2O_3 carrier	E.I. du Pont de Nemours	CO	2000 ppm NO, 10,000 ppm CO, 40,000/hr (1) 340°C, (2) 380°C, (3) 550°C.
Oxides of Cu, Cr and Fe on Al_2O_3	Agency of Industrial Sciences and Technologies.	NH_3	I 300 ppm NO, 300 ppm SO_2 300 ppm NH_3, 2.5% O_2, 15,000/hr, 350°C. (1) 0 hr, (2) 4 hrs, (3) 12 hrs. II. 300 ppm NO, 300 ppm NH_3 2.5% O_2, 10.2% H_2O 15,000/hr, 320–480°C.
$V_{12-x-y}Mo_xW_yAl_z$ $x=0-8$, $y=0-5$, $x+y= 0.3-8$, $z= 40-480$	Sumitomo Chemical Co., Ltd.	NH_3	130-180 ppm NO_x, 800-1200 ppm SO_2, 50-100 ppm SO_3, 1.3/1 NH_3/NO_x, 5000-14000/hr, (1) 250°, (2) 350°C
CuO on γ-Al_2O_3	Asahi Chemical Industry Co., Ltd.	NH_3	230 ppm NO, 2.5% O_2, 10% CO_2, 10% H_2O, 210 ppm NH_3. 12,500/hr, 200-500°C
CuO on mordenite	Toray Industries, Inc.	NH_3	300 ppm NO 300 ppm SO_2, 300 ppm NH_3, 2% O_2, 9% H_2O, 1000 ml/min. (1) 275°, (2) 300°C, (3) 350°C.
MnO_2	Nippon Steel Corp.	NH_3	I. 873 ppm NO_x, NH_3/NO_x ratio =2.1, 200°C, 4536/hr II. 830 ppm NO_x, NH_3/NO_x = 2.05, 180°C, 4536/hr.
1-10% CuO and 1-10% oxides of Zn, Mn and Fe on Al_2O_3	Idemitsu Kosan Co., Ltd.	NH_3	498 ppm NO_x, 1005 ppm SO_2 5% O_2, 10.5% H_2O, 500 ppm NH_3, 10^4/hr, 350°-400°C.
Cu oxide on Al_2O_3	Asahi Chemical Industry Co., Ltd.	NH_3	200 ppm NO_x, 580 ppm SO_x 2.6% O_2, 210 ppm NH_3, 1 m^3/hr 350°C (1) 1 (2) 10 (3) 30 days.

Conversion or Outlet Conc.	Selective	SO_2 resistant	Duration	Ref. No.	Remarks
(1) 25% (2) 50% (3) 90%	no	NR	NR	439	A=metal with atomic no. 11-51, or 89-103. B= part Pt group metal, part non-Pt metal, ionic radius 0.4-1.4A, 25% multivalent metal in 1 oxide state, $\geq$5% same metal in different oxide state.
I. (1) 69.0% (2) 97.1% (3) 99-100% II. ~100%	yes	yes	NR	441	Example: 10% CuO on Al_2O_3.
(1) 68-74 % (2) 94-99 %	yes	yes	1150 hr	442	α - Al_2O_3 carrier. Example: $V_9Mo_3Al_{120}$ All cations present as oxides.
NR	yes	NR	NR	451	Superior catalytic activity and durability to CuO prepared from nitrates.
(1) 80.8% (2) 93.7% (3) 98.8%	yes	yes	NR	452	Pretreat mordenite with NH_4^+ solution. 3.1% Cu.
I. 20 ppm II. 31 ppm	yes	NR	NR	453	
95%	yes	yes	NR	455	
(1) 92% (2) 90%, (3)88%	yes	yes	See left and remarks	457	Heat catalyst to 480°C for 2 hrs on 31st day, activity restored. (cont.)

(cont.) Catalyst	Company/Institution	Reductant	Operating Conditions
V, Cr, Mo and/or W on θ-or γ-Al_2O_3	Toray Industries, Inc.	NH_3	300 ppm NO, 300 ppm SO_2, 200 ppm NH_3, 9% H_2O, 2% O_2, 10^4/hr, 250°C.
NH_3	Hitachi, Ltd; Babcock, Hitachi K.K.	NH_3	300 ppm NO_x, 500 ppm SO_2, 30 ppm SO_3, 50 mg/m^3 dust, 10% H_2O, 1.5% O_2, 11% CO_2, 1.1 NH_3/NO, 10,000/hr, (100,000 m^3/hr flow, 370-420°C.
V and/or Fe on θ - or K-Al_2O_3 carrier	Toray Industries, Inc.	NH_3	300 ppm NO, 300 ppm SO_2, 300 ppm NH_3. 9% H_2O, 2% O_2, 5000/hr, 275°C.
Cu and/or Ni porous C fiber	Toray Industries, Inc.	CO	0.5% O_2, 0.2% NO, 1.5% CO, 10^4/hr, 400°C.
Vanadium sulfate on Al_2O_3	Mitsui Petrochemical Industries, Ltd.	NH_3	300 ppm NO, 3% O_2, 15% CO_2, 12 12% H_2O, 300 ppm SO_2, 300 ppm NH_3, 15,000/hr, 350°C.
MnO_2 on Ti grid carrier	Nippon Steel Corp.	NH_3	200 ppm NO, 200 ppm NH_3, 13% O_2, 2 x 10^4/hr, 150-300° C.
V_2O_5 on Al_2O_3	Nippon Steel Corp.	NH_3	300 ppm NO, 1000 ppm SO_2, 400 ppm NH_3, 15% O_2, 2 ℓ/min, 200°C.
Zeolite-type Aluminum silicate	Chiyoda Chemical Engineering and Construction Co., Ltd	NH_3	300 ppm NO, 2% H_2O, 15% O_2, 120 ℓ/hr, 300°C.
CuO-Al_2O_3	Niigata Engr. Co., Ltd.	NH_3	5% O_2, 10% H_2O, 400 ppm NO, 200 ppm SO_2, 400 ppm NH_3, 2000/hr 340-480°C.
Ni, Cu, Th and/or La on mullite	Ube Industries Ltd.	CO/H_2	0.25% NO, 1.0% H_2, 1.7% CO, 0.4% O_2, 0.03% C_2H_4, 8% CO_2, 3.5% H_2O, 40 ppm SO_2, balance He; 65,000/hr. (1) 1hr, (2) 2 hrs, (3) 5 hrs.
$FeSO_4$ on γ-Al_2O_3, C, SiO_2 or diatomaceous earth	Japan Gasoline Co. Ltd.	NH_3	500 ppm NO, 500 ppm NH_3, 1500 ppm SO_2, 3% O_2, 10% H_2O, 30-500 ℓ/hr, 550°C.

Conversion or Outlet Conc.	Selective	SO_2 Resistant	Duration	Ref. No.	Remarks
99.5%	yes	yes	NR	458	
>90%	yes	yes	4000 hr	459	Treatment was upstream of economizer. Treat gas after economizer at 360°C, conversion was only 60% after 1000 hrs.
93.7%	yes	yes	NR	460	
90%	no	NR	NR	461	C fibers sensitized in $SnCl_2$ and HCl, activate in $PdCl_2$ + HCl, then dipped in $CuSO_4$
99%	yes	yes	NR	462	3.9% V, $SO_4/V = 1$
>95%	yes	NR	NR	464	
$\geq$ 99%	yes	yes	140 hrs	465	Catalyst pretreated with 2000 ppm SO_2 at 2000°C
25 ppm	yes	NR	NR	466	NH_3 was provided by pretreatment of "catalyst" with NH_4NO_3
92.2%	yes	yes	NR	467	
(1) 96% (2) 94% (3) 88%	no	yes	NR	468	Example: Ni:Cu:Th = 0.7:0.3:0.8; 8% Ni + Cu.
98%	yes	yes	NR	469	